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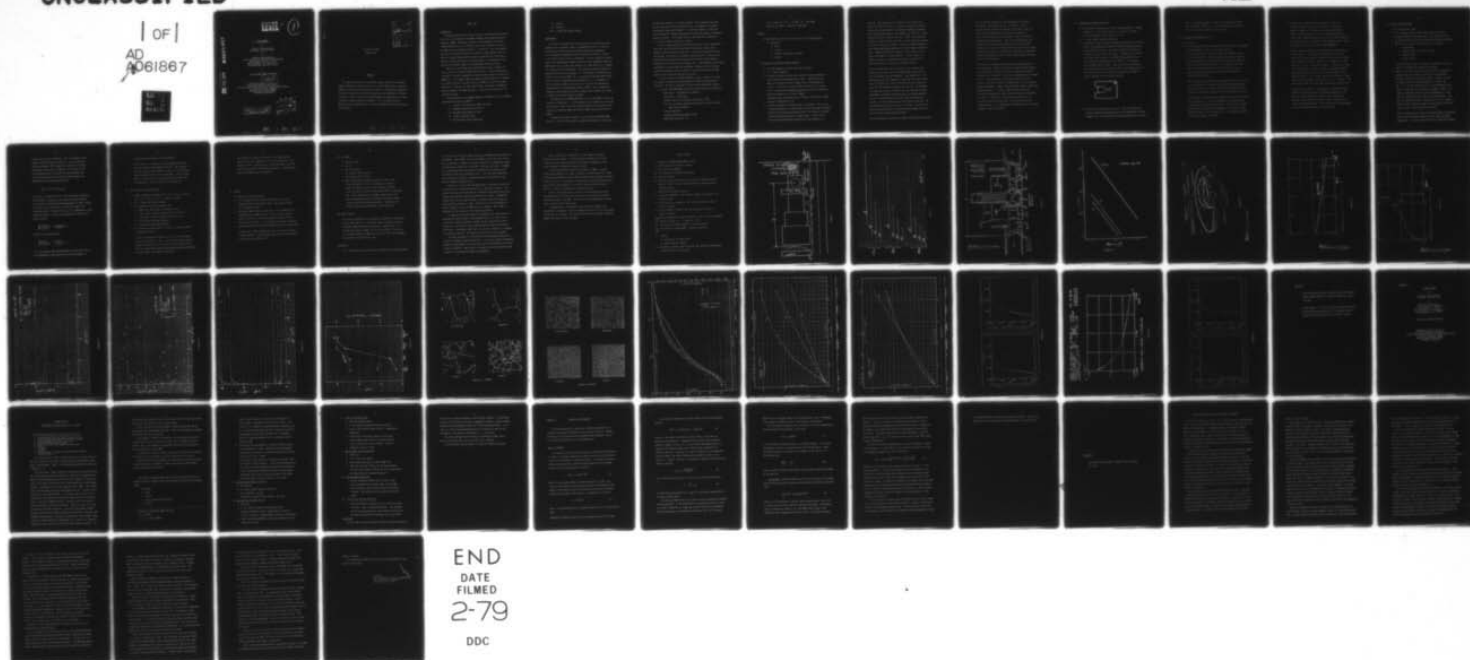
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FEASIBILITY STUDY ON THE USE OF SMALL-ANGLE NEUTRON SCATTERING --ETC(U)
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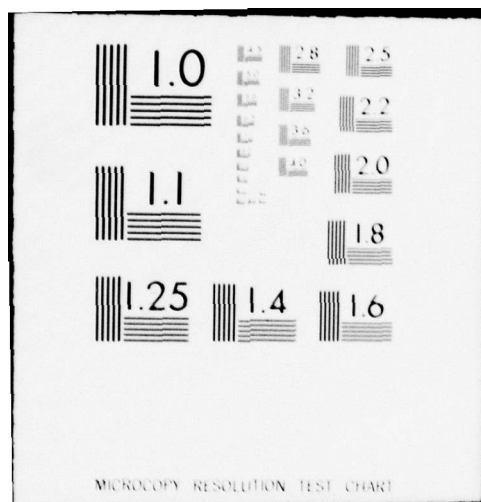
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9 Final Report

to

Office of Naval Research
Arlington, Virginia 22217

by

10 Professor Herbert/Herman
State University of New York at Stony Brook
Stony Brook, New York 11794
(Mailing Address - 11 Meroke Trail
Port Jefferson, New York 11777)

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11 1978

6 Feasibility Study on the Use of
Small-Angle Neutron Scattering for
Microstructural Determinations of Technological Alloys,
Carried out at Atomic Weapons
Research Establishment (AWRE),
Aldermaston, England

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Feasibility Study

SANS for NDE

ABSTRACT

A study was carried out at the Atomic Weapons Research Establishment (AWRE), Aldermaston, England, on the use of small-angle neutron scattering (SANS) for non-destructive evaluation of alloys of technological importance. A range of ferrous and nickel-based superalloys were examined in different conditions. SANS is sensitive to the development of heterogeneities having sufficient scattering power for neutrons. It is concluded that this method can play an important role in alloy development and long range alloy test programs.

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SANS - NDE

Introduction

During the month of July, 1978, a series of experiments were carried out by this investigator at the HERALD REACTOR, Atomic Weapons Research Laboratory (AWRE), Aldermaston, England. The program was carried out cooperatively with Dr. R.J.R. Miller, who is the scientific director of the SANS facility, together with Mr. A.J. Hitchcock. This investigator's aspect of the study was funded by the Office of Naval Research and the Naval Air Systems Command and was aimed at examining the feasibility of employing small-angle neutron scattering (SANS) as a technique for non-destructive evaluation (NDE) of technologically important alloys.

Previously, such studies were performed principally by workers in Italy as well as at other European SANS facilities. These programs were reviewed in a previous comprehensive report produced under the auspices of NAVAIR (1). Last summer's AWRE experimental program enabled us to examine, on a first-hand basis, the possibilities of SANS as a tool for the NDE specialist. There are, in fact, some definite conclusions forthcoming which are based on the AWRE experience. These will be discussed in the following sections.

As was outlined in the Progress Report (2) (Appendix 1), the following experimental program was planned:

- I. Fatigue of Ti-Alloy Disks (Webs and Rims).
- II. Hydrogen Embrittlement of HY-130.
- III. Hydrogen Embrittlement of Iron.
- IV. Creep of Stainless Steel.
- V. Hot-Isostatic Pressed Superalloy.

- VI. Incoloy.
- VII. Hastelloy.
- VIII. Inconel and Alumina-Compacts.

Experimental

Portions of the above goals were carried out at the Institut Laue-Langevin (ILL), Grenoble, France, in cooperation with Dr. G. Kostorz, with whom this writer has had an ongoing collaborative agreement. Dr. M. Suenaga of Brookhaven National Laboratory assisted Dr. Kostorz in the carrying out of the ILL experiments. In fact, during the performance of this experimental program, there was close cooperation among the AWRE people (e.g., my host and co-worker, Dr. Robin Miller), Dr. Suenaga, Dr. Kostorz and this investigator. We frequently spoke by telephone on matters relating to the experiments. If, for example, the intensity was found to be too low at AWRE (e.g., to perform the long wavelength experiments on Ti), the experiment would be moved to ILL. Dr. Suenaga, in fact, travelled to AWRE on his way to ILL to pick up the appropriate specimens which had already been studied at AWRE. Thus, a comparison becomes possible between the results obtained from a medium flux reactor (~ 5 MW) and a high flux reactor (~ 50 MW).

The details of the HERALD REACTOR, as well as the SANS facility, are discussed in Reference 1. It will, however, be of interest to review the facility here, since it has just recently become fully operational, and, actually, our work was among the earliest SANS projects carried out on HERALD.

Schematically shown in Figure 1 is a side view of the AWRE SANS facility. For all of our studies a specimen-to-detector, D , spacing of

4.3 meter was employed. The vacuum chambers in the neutron flight path are on tracks, and, using an overhead crane, the chambers can be added and removed, changing the length D. The specimen chamber is large and easily accessible. Neutron wavelengths are determined through a mechanical velocity selector. The Q-ranges which are available for D spacings of 1.7, 3 and 4.3 meters are given in Figure 2.

The LETI area detector was installed in late 1977 and is operational. There were some difficulties experienced from spurious counts, most probably, due to electronic connections. It was thus required to process the raw data "by hand", using computer-assisted methods. There was also some difficulty encountered in transferring the data from the detector storage unit to the disks in the computer room. These temporary difficulties contributed to slowing down the experiments. However, due to excellent technical assistance, and as a result of fine cooperation with the AWRE people, these limitations were not too serious.

The cold source of the HERALD REACTOR is shown in Figure 3, and the flux output is plotted versus wavelength for different refrigerants in Figure 4a. The LETI is depicted in Figure 4b (after RJR Miller, AWRE).

In review, the HERALD-SANS facility has the following specifications:

Power level: 5 MW (Medium flux Reactor)

Cold source: liquid H_2/D_2

Cold Be filter for D high pass filtration ($< 4 \text{ \AA}$).

Fine slit collimator coupled with mechanical velocity selector
(Mg-Cd alloy cylinder).

LETI area detector

Incident neutron wavelength; 4-25 \AA .

Wavelength resolution: 8%

Flux at sample at 6 \AA : $2 \times 10^2 \text{ mm}^{-2} \text{ sec}^{-1}$ (at 5 MW)

Beam size at sample: 30mm wide x 20mm high

Results

The experimental results will be reported in the following manner:

- I. Experiment
 - A. Alloy
 - B. Contact
 - C. Types of Experiments performed
 - D. Results

I. Fatigue of Ti-Alloy Disks (Webs and Rims)

- A. Ti-6V-4Al (Samples cut from rotary disk forgings)
- B. Dr. G. London, NAVAIR
- C. A range of fatigue specimens were examined, having undergone low cycle fatigue at room temperature or 300°F. SANS spectra were taken at the reduced section (middle) and at the unloaded end section. In one case, a run was made near the tip of a fatigue crack. (Cold neutron radiography experiments were also performed; under the auspices of G. Tuckey of AWRE.)

Experiments were carried out with 6.5 \AA and 8.2 \AA neutrons at AWRE and with 12 \AA neutrons at ILL.
- D. For these specimens a difference plot is considered. That is, the scattered intensity, at a given angle, between the middle (fatigued) section and the end (non-fatigued) section. An increase in scattering has been detected from the loaded section. This is due to either fatigue-induced phase modification, void formation, or

cracking. TEM studies must be carried out concurrently with SANS to delineate this effect. Figure 5a-b show the scattering spectra for end and middle of the fatigue specimen, the Guinier plot, and the Porod analysis (see Appendix 2). There is a linear region which gives a Guinier radius of $\sim 200 \text{ \AA}$. The Porod plot indicates a complex surface structure, but does imply the presence of discrete particles (or voids). The slope could be changed to zero through appropriate additions of surface-induced scattering terms. Nothing particularly definitive can be stated at this time on the matter of a Porod analysis. Ideally, however, it should be possible to determine the specific surface of the scattering entities, and, thus, the volume fraction.

We were concerned that the Ti_3Al phase, which is known to be present in this alloy, was giving double-Bragg scattering for the 6 \AA neutrons which were employed. There should be a 7 and 9 \AA spacing within the compound. A study was thus also carried out at 8.2 \AA and 12 \AA . The 8.2 \AA results were very similar to those for 6 \AA , though the intensity was very low. We attempted to examine the system at 12 \AA , but the time required for proper experimental statistics would have taken up an entire weekend, so only a relatively short run at the longer wavelength was feasible. No clear changes for the 12 \AA run were detected. The specimen was then sent to ILL for a 12 \AA study. The results of that study indicate that no significant double-Bragg scattering exists for this system following fatigue.

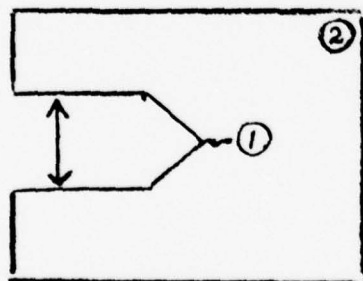
The rim and web of the disks are known to have different textures,

but this was not reflected in the development of scattering anisotropy. The implications are: 1) that the α' -phase (Ti_3Al), which is known to be present in this alloy, does not have a strong orientational relation with the matrix; or 2) the scattering which we are observing is due to voids forming within the loaded region. Since the long wavelength runs yielded nothing significantly different from the lower wavelength runs, it could be concluded that the voids are the sources of scattering (Ti_3Al has a sufficiently large Bragg spacing which should yield measurable intensity due to double-Bragg scattering, even for 8.2 \AA neutrons).

It must thus be concluded that the present SANS experiments on the rotary disks, though not dramatically showing fatigue damage, are sufficiently sensitive to the cyclic loading that further investigation is warranted. Cold neutron radiography was performed as well. The availability of low energy neutrons from the reactor's cold source presents the opportunity of high resolution neutron radiography. Indeed, some preliminary studies dramatically demonstrated this. G. Tuckey of AWRE examined the cracked specimen and compared the result with normal thermal radiography. The result is surprising, in that using cold neutrons it is possible to detect the crack when it was barely visible with thermal neutrons. It is hoped that a Navy-AWRE contact can follow-through on some joint studies.

II. Hydrogen Embrittlement of HY-130

- A. HY-130 Steel (Pressure vessel steel, tough and weldable: nominal composition - C (0.10%); Mn (0.75%); Si (0.25%); Cr (0.50%); Ni (5%); Mo (0.50%); Some V and Al (< 0.1%).
- B. H. Zannis, NSRDC, Annapolis, Maryland.
- C. To examine in situ electrolytic charging of hydrogen into 1/8 inch plate. The specimens were notched at NSRDC and remained loaded by the insertion of a peg. The notch strength of the material indicates a K_{CI} of 70 ksi root inches. Charging will be carried out at the target position on the AWRE SANS spectrometer in a NaCl solution, potentiostatically at 1000 mV. Two specimens were received from Mr. Zannis. Base lines will be established (at the notch tip and away from the crack) See drawing below for positions (1) and (2), to ascertain the potential for SANS to detect incipient stress corrosion cracking.



- D. No results are available at this time. There was insufficient time to perform the experiments during this investigator's period at AWRE. It is to be noted that these experiments must be carried

out in a saturation magnetic field to avoid troublesome domain wall scattering. Miller et al. will carry out these investigations and report the results to the U.S. Navy.

III. Hydrogen Embrittlement of Iron

- A. Ferrovac E
- B. Dr. M. Suenaga, Director, Metallurgy & Materials Science, Brookhaven National Laboratory, Upton, Long Island, New York.
- C. It is known that iron is subject to attack by hydrogen, giving rise to embrittlement. TEM has limited usefulness for examining the initial aspects of this embrittlement due to difficulties in obtaining valid particle statistics. Using SANS, on the other hand, it is possible to employ a bulk specimen and, thus, to examine the average properties associated with hydrogen (or methane) bubble formation.

These experiments were carried out cooperatively with Dr. G. Kostorz at ILL and with Dr. M. Suenaga of Brookhaven National Laboratory. The ILL facility has the intensity and computer hardware which is required for scattering from "pure" metals and alloys with defects.

The specimens to be studied were charged at 900 psi with H_2 at $475^\circ C$ for different times. The D11a spectrometer was employed at the 20 meter position using 9.84 \AA neutrons. Of course, during SANS measurements, the specimens were magnetically saturated (placed between the poles of an electromagnet of 9.5 kilooersted field) to avoid magnetic domain scattering.

- D. Porod plots are shown on Figures 6a and 6b for pure iron specimens differing in grain size. There is a significant amount of scatter in this data, which could be decreased if longer scattering times were used: 15 minutes was generally used in these experiments, whereas an hour or two would have been preferred (But time is frequently an unobtainable luxury in these experiments, due to the very busy reactor schedule)..

However, several interesting facts emerge from a Porod analysis of these results. Most apparent from the plots is the fact that the IQ^4 saturation level (nominally level region), increases with an increase in time of charging (i.e., compare for Fe "A", Figure 6a, $I(70\text{ h}) - I(\text{As})$ with $I(40\text{ h}) - I(\text{As})$). Also, there is no strong grain size effect, implying that bubble formation (believed to be CH_4) does not occur at grain boundaries, but within the grains. It is further important to note that a linear region was not observed in any of the $\ln I$ - vs. - Q^2 plots. This implies the presence of a very wide distribution in bubble size.

Further work is required in this program to delineate the Porod behavior with greater accuracy, and (though difficult in this case) to attempt comparisons with TEM. What might be more fruitful, on the other hand, would be fracture studies, correlated with SANS measurements. These results give us encouragement for the HY-130 studies, which are now under way.

IV. Creep of Stainless Steel

- A. Type 304 Stainless Steel
- B. Dr. M. Suenaga, Director, Metallurgy & Materials Science,
Brookhaven National Laboratory, Upton, Long Island, New York.
- C. The following type 304 stainless steels specimens were studied at
AWRE and at ILL (under the auspices of Dr. G. Kostorz):
 - 1. As-received
 - 2. Soaked at 576°C for 144 h, no load.
 - 3. Crept at 576°C
 - 4. Crept at 576°C

Since the stainless steel in question is austenitic it was not required to magnetically saturate the specimen.

- D. Again, as for the Ferrovac E specimens which were charged with hydrogen, the crept stainless (as well as the as-received materials) showed no clear Guinier region. However, as shown in Figures 7a and 7b, the Porod behavior is classic, the IQ^4 - vs. - Q^4 plots levelling off at a value dependent on the amount of creep that the specimen has undergone. Creep 1 and 2 refer to different times of loading at-temperature. These times are currently not available, and are being sought. However, the results are clear: Annealing will give an increase in effective total scattering volume, and annealing at 576°C with load will give a rather significant increase.

Shown in Figure 7c-d are optical micrographs taken of these specimens in the as-received, annealed, and two crept conditions, respectively. It can be seen that the major change which appears

during creep is grain refinement. This is reflected in both hardness and IQ^4 increases, Figure 7b. In fact, the Porod analysis parameter, IQ^4 increases by a factor of 8 from the as-received to the creep 1 condition. On the basis of this large change from the as-received to crept condition, one can calculate the ratio of scattering surface between two conditions from the Porod approximation:

$$\frac{d\sigma}{d\Omega} (I) \times Q^4 = 2\pi S (C_a - C_b)^2$$

for $QR_g \gtrsim 2.5$; where $d\sigma/d\Omega (I)$ is the differential scattering cross-section, C_a and C_b are the scattering length densities for the matrix and scattering center (analogous to atomic scattering factors for x-rays), and S is the surface area of the scattering volume. Assuming that $(C_a - C_b)$ is independent of alloy condition (which is a good just approximation), ratios can be taken, yielding:

$$\frac{IQ^4 (\text{creep 1})}{IQ^4 (\text{as-rec})} = \frac{S (\text{creep 1})}{S (\text{as-rec})} = 8$$

and for the annealed specimen:

$$\frac{IQ^4 (\text{ann.})}{IQ^4 (\text{as-rec.})} = \frac{S (\text{ann.})}{S (\text{as-rec.})} = 2$$

It is thus obvious that annealing under load will give rise to the development of approximately four times the number of

scattering centers than for no-load annealing.

At this time, it is difficult to conclude what will be the cause of increase in both hardness and IQ^4 . These changes will arise either from a decrease in grain size or from load-induced precipitation of a fine second phase. Also of importance is the possibility of forming micro-voids under-load, at-temperature. Clearly, TEM studies are called for.

V. Hot-Isostatic Pressed Superalloy

- A. UDIMET 700 (Nominal composition; Ni, 53%; Cr, 15%; Co, 18.5%; Mo, 5.2%; Al, 4.3%; Ti, 3.5%; C, 0.08%; B, 0.030%).
- B. Dr. V. Wilms; MTU; Munich, Germany.
- C. Heat treatment and aging of Hipped alloy:
 - 1. UDIMET 700: HIP @ 1100°C, 1050 Bar for 2 hrs.
 - 2. U-700: (HIP), Solution treated at 1175°C for 2 hrs.
 - 3. U-700 (HIP), Solution treated at 1120°C for 2 hrs.
 - 4. #2 plus three-stage age-hardening.
 - 5. #3 plus three-stage age-hardening.
- D. The resulting scattering data for series 1, 2 and 4 are given in Figures 8a, b and c.

For the aged specimen #4, Figure 8a, one sees a clear Guinier region in the range $1.5 < RgQ < 3.1$, the slope of which yields a Guinier radius of 61 \AA . As can also be seen from the plots, there are linear Porod regions, which then deviate from linearity, due, most likely, to incoherent scattering.

The 61 \AA^0 particles which are observed in the aged specimen (#4, Figure 8a) are comparable in size with precipitates found normally in TEM. Clearly, there is scattering which is arising from internal surfaces other than precipitates. A comprehensive combined TEM-SANS study is appropriate.

VI. Incoloy

- A. 800 Alloy (Fracture specimen)
- B. Dr. M. Suenaga, Director, Metallurgy & Materials Science,
Brookhaven National Laboratory, Upton, Long Island, New York.
- C. Stress ruptured specimen
- D. Preliminary AWRE results are uncertain: This specimen will be examined further by AWRE workers. The ILL study is shown in Figure 9a for end vs. tip (the fractured portion) for a thin sheet particle analysis. It is seen that the radius of gyration increases about 10% from the non-deformed end to the fracture tip.

In Figure 9b is shown a Guinier plot for this alloy in solution treated condition. A low Q region shows an R_g of 591 \AA^0 , whereas a high Q analysis yields 170 \AA^0 .

VII. Hastelloy

- A. Alloy X (Disks)
- B. See VI.
- C. #1; as-received.
#2; soaked at 700°C, no load.
#3; crept at 700°C (14 ksi).
- D. Good scattering, but nothing significant occurs on the scattering profile. A flat disk analysis is shown in Figure 10, where the linear region indicates a Guinier radius of 50 Å for solution treated and 42 Å for the deformed tip. These similarities are somewhat interesting, since optical metallography shows significant differences between solution treated and deformed specimens. TEM studies are currently under way at Brookhaven on these specimens.

VIII. Miscellaneous:

The Inconel experiments (Pratt & Whitney, Palm Beach, Florida) have not yet been carried out. The porous alumina specimens have been examined at AWRE during this investigator's tenure there. The transmission factors, however, are not yet available. These tests are currently underway at AWRE, and the final results will hopefully be available by the end of this year.

Conclusions

It is clear that only a fraction was completed of our intended program.

The initial goal of this one month study was to demonstrate the feasibility of SANS-NDE. And, indeed, in this investigator's view, this was, in the main, accomplished. However, one month's research on a new machine cannot be sufficient to accomplish a great deal in the way of results. A number of the experiments which are presently reported on were carried out by colleagues at the D11a spectrometer at ILL. This was necessitated both due to an effort to obtain a reasonable amount of data and due to the greater intensity available at ILL.

Overall, it is clear that the SANS method is particularly sensitive to the presence of a high void density (e.g., hydrogen in metals). The presence of voids can be detected at a relatively low volume percent (and further work should be aimed at determining this figure). Deformation-induced voids are also good scatterers, and thus creep and fatigue are good candidate processes for SANS. Voids due to less-than-theoretical-density compacts can also be detected (e.g., alumina). Thus, SANS is employable in HIP studies of, for example, superalloys.

As shown by the Italian workers (see Reference 1), second phases in technological alloys (steels, aluminum-base alloys, etc.) give rise to significant scattered intensity in SANS, and the coarsening of second phases (e.g., in service) can be readily detected (without thinning for TEM study!). Furthermore, SANS "sees" the bulk and, therefore, is expected to average the material. Good particle statistics are thus expected.

Of great importance is the occurrence of double-Bragg scattering in studies of deformed crystalline materials. This can be avoided by employing neutrons of wavelengths of at least twice the largest Bragg spacing for the crystal in question. Some concern, for example, arose in the present work relative to Ti_3Al which has large cell parameters.

Thus a cold source is required for a large number of physical metallurgical-type studies. A further benefit of having a cold source is the ability to carry out NDE with cold neutron radiography, a most useful technique for looking at complex systems.

In discussion with Dr. Miller and co-workers at AWRE, it became apparent that SANS will not always be useful, but rather there are a range of interesting problems for which SANS could make highly important contributions (e.g., Ref. 1). We agreed, in particular, that solid solution alloys per se offer little prospects for acting as strong scattering systems. Unless, of course, considerable void formation occurs.

Rather, for the most part, it will be the multiphase systems and the modification of the phase distribution in-service that will present interesting studies using SANS. This fact has been demonstrated time and again, here and in the Italian work.

The AWRE group is continuing to carry out and to complete the experiments listed above. We are in agreement that this sort of work should by all means be continued. This writer shall present a proposal to ONR and NAVAIR such that it can be continued.

Figure Captions

1. Schematic of HERALD REACTOR SANS facility.
2. Q (\AA^{-1}) ranges for different D values.
3. Cold source within HERALD.
- 4a. Flux spectrum for different refrigerants.
- 4b. LETI Area Detector.
- 5a. Difference scattering spectra in the form of a Guinier plot (See Appendix 2) for Ti-alloy. Difference between fatigued and non-fatigued regions.
- b. Porod plot of difference spectra.
6. Porod plots (Appendix 2) for hydrogen charged into Ferrovac E for different times.
- 7a. Porod Analysis (see Appendix 2) for type 304 stainless steel in various conditions.
- 7b. IQ^4 and Rockwell A hardness for type 304 stainless steel in various conditions.
- 7c. Optical micrographs of type 304 stainless steel in various conditions, and
- 7d. related to Figure 7b. 7c - 1000x; 7d - 100 x.
- 8a. Guinier plot for hipped UDIMET in various conditions.
- 8b. Porod plots for hipped UDIMET in various conditions.
- 8c. and
9. Incoloy:
 - a. Guinier plot for thin sheets.
 - b. Guinier plot for spheres.
10. Hastelloy: Bottom, solution treated; top, deformed at-temperature. Guinier analysis for thin sheet.

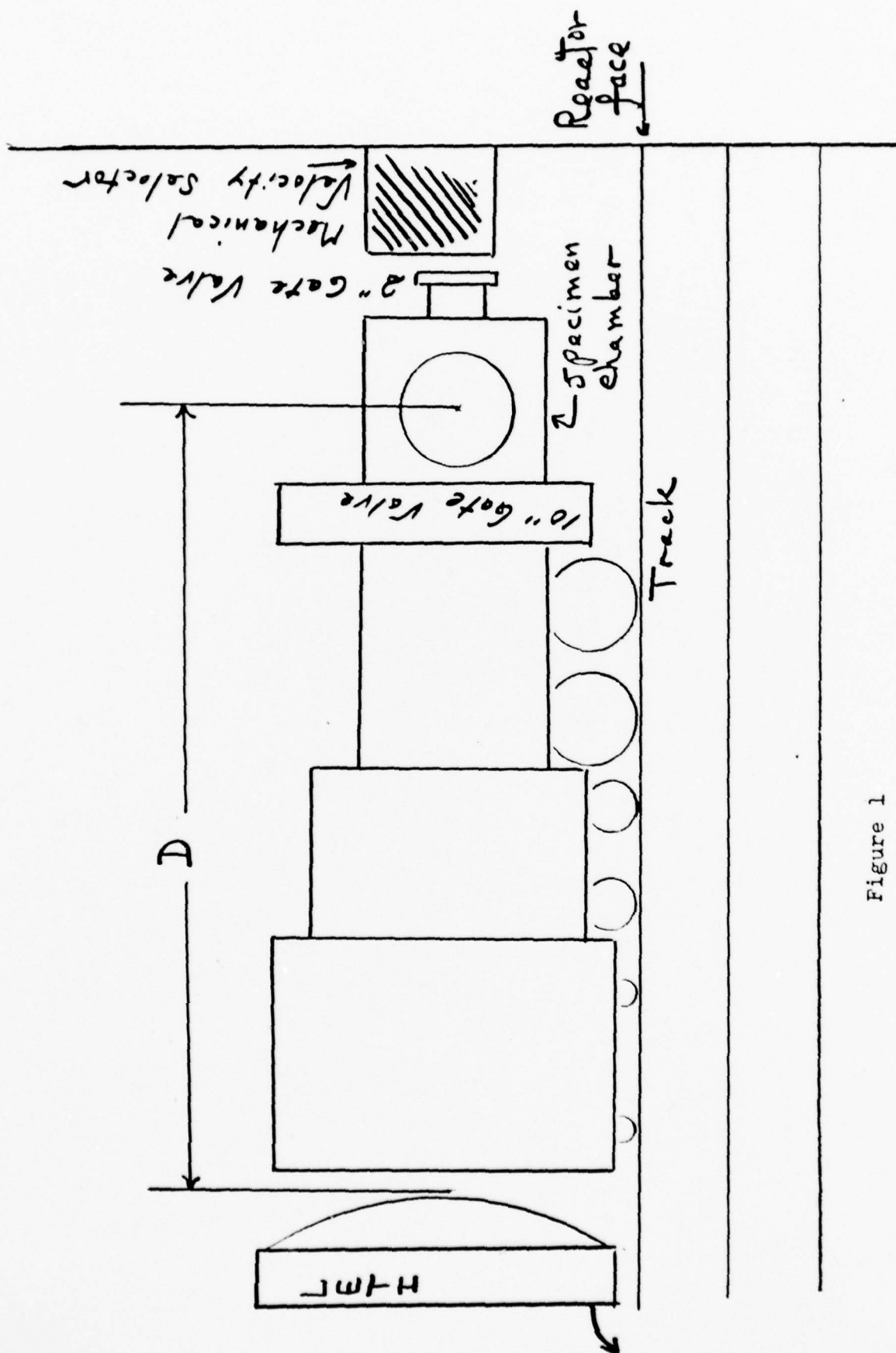


Figure 1

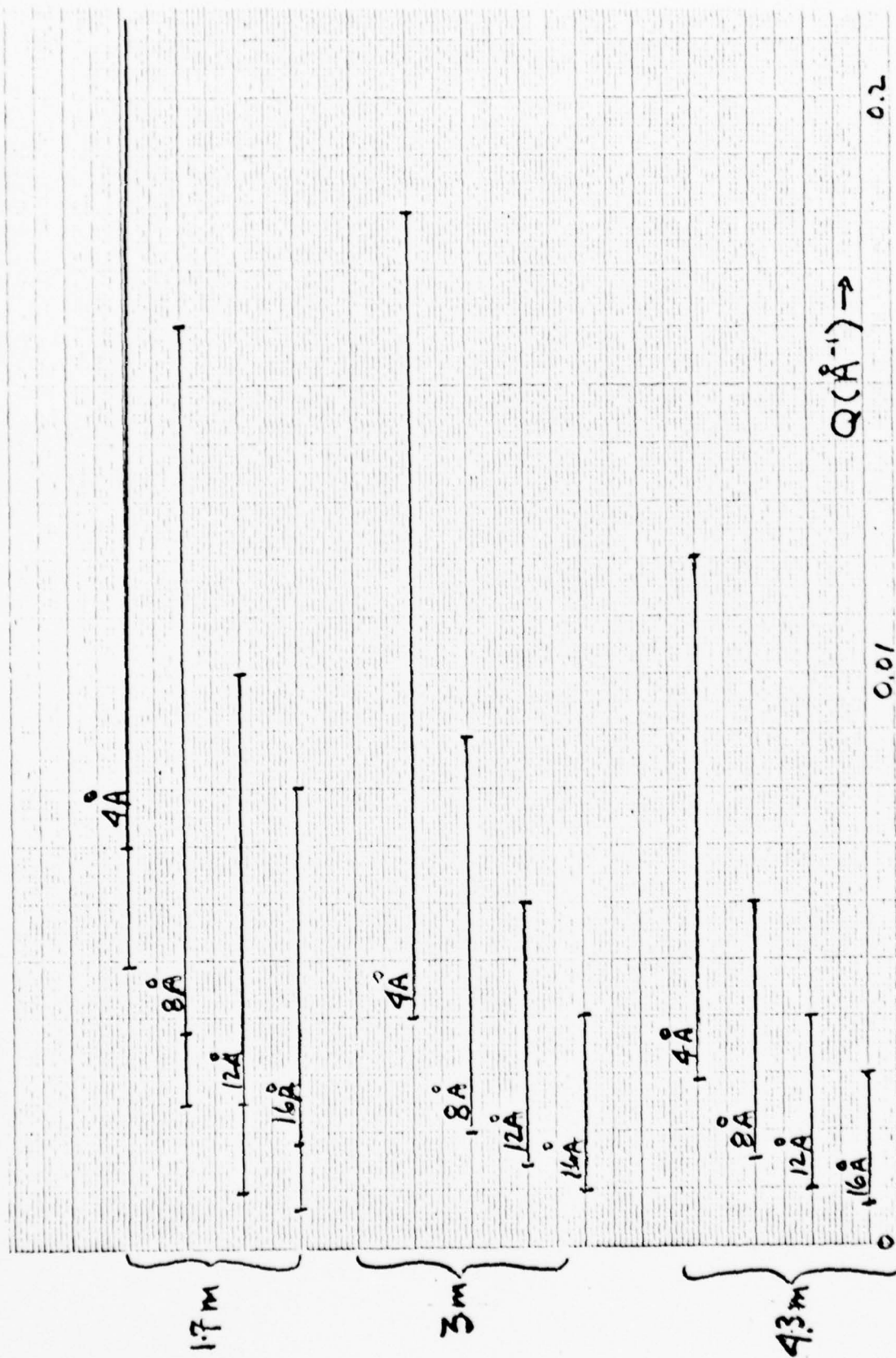


Figure 2

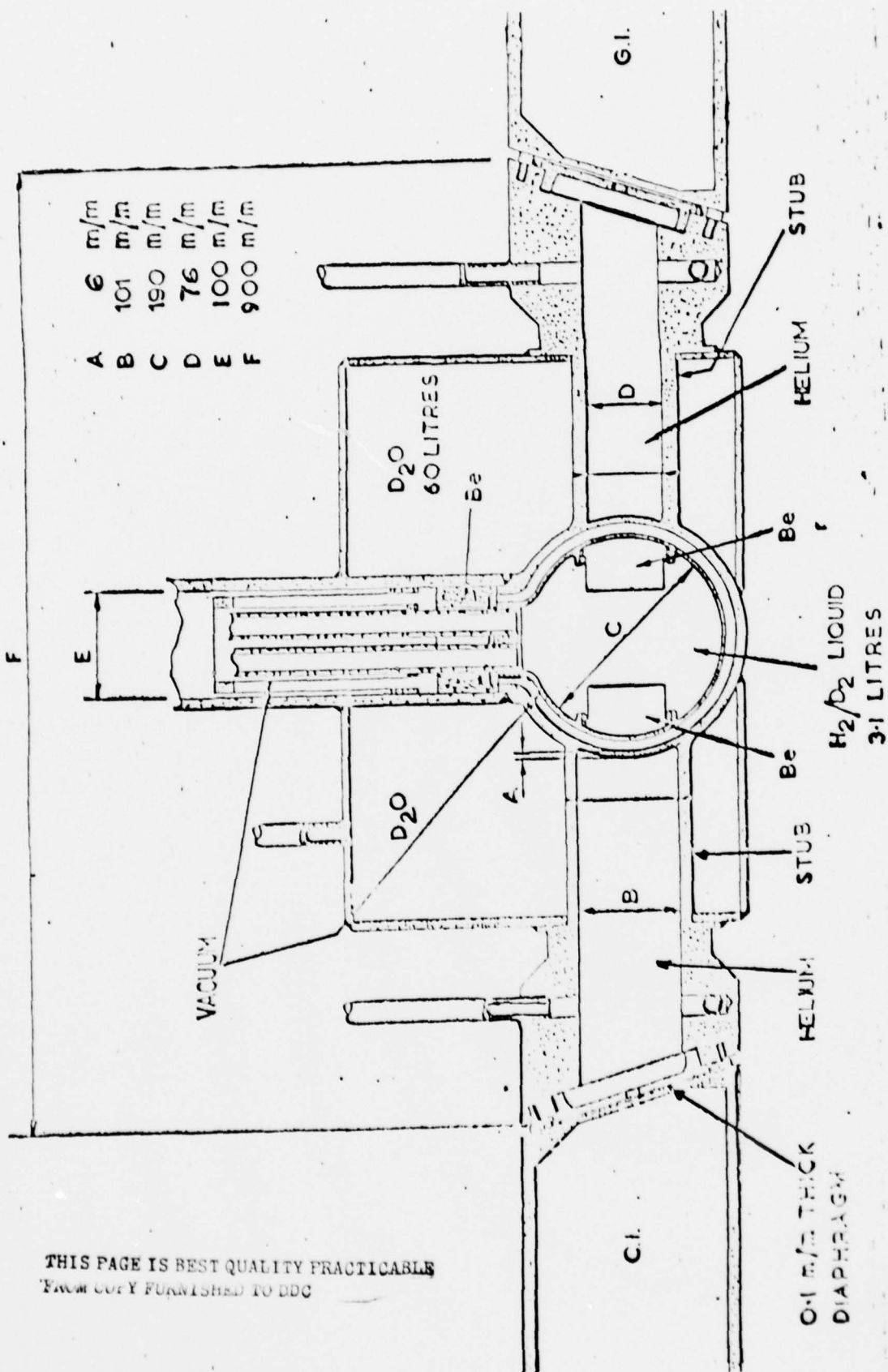


Figure 3

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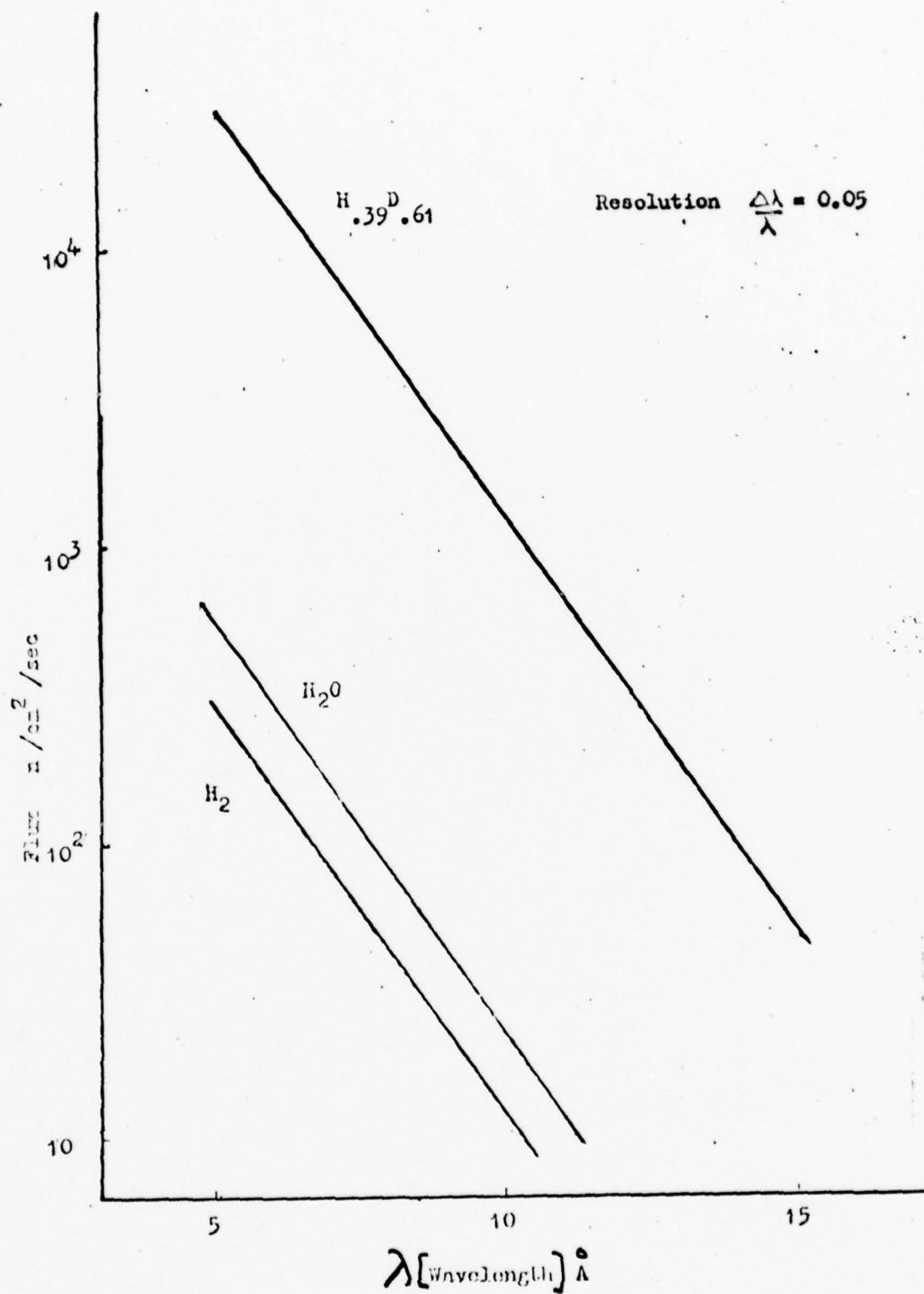


Figure 4a

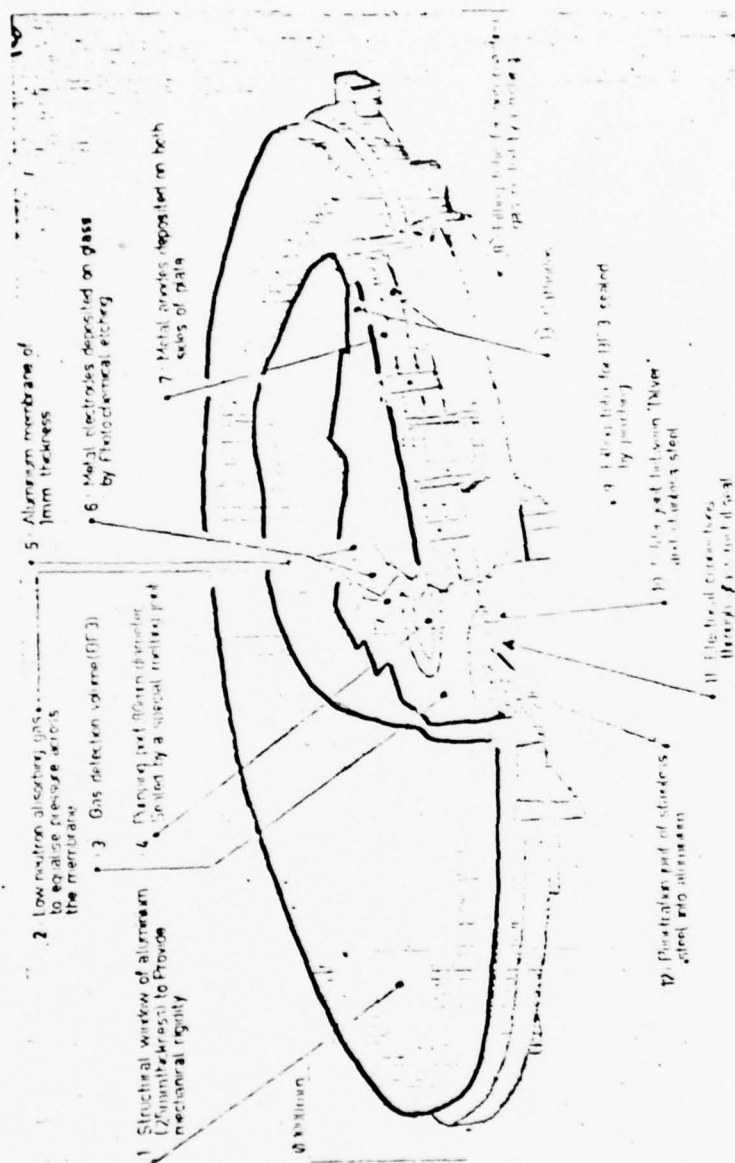


Figure 4b

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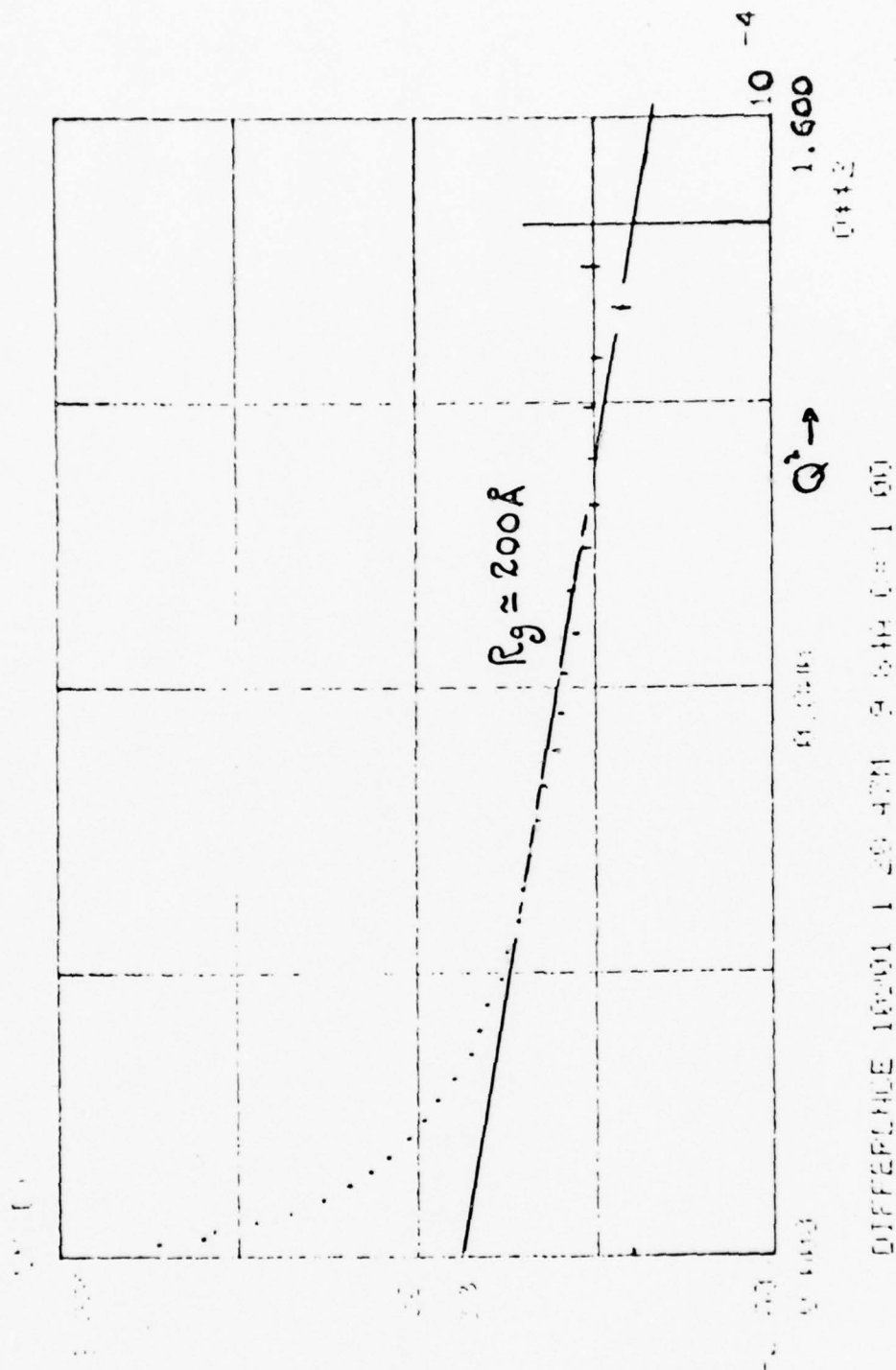


Figure 5a

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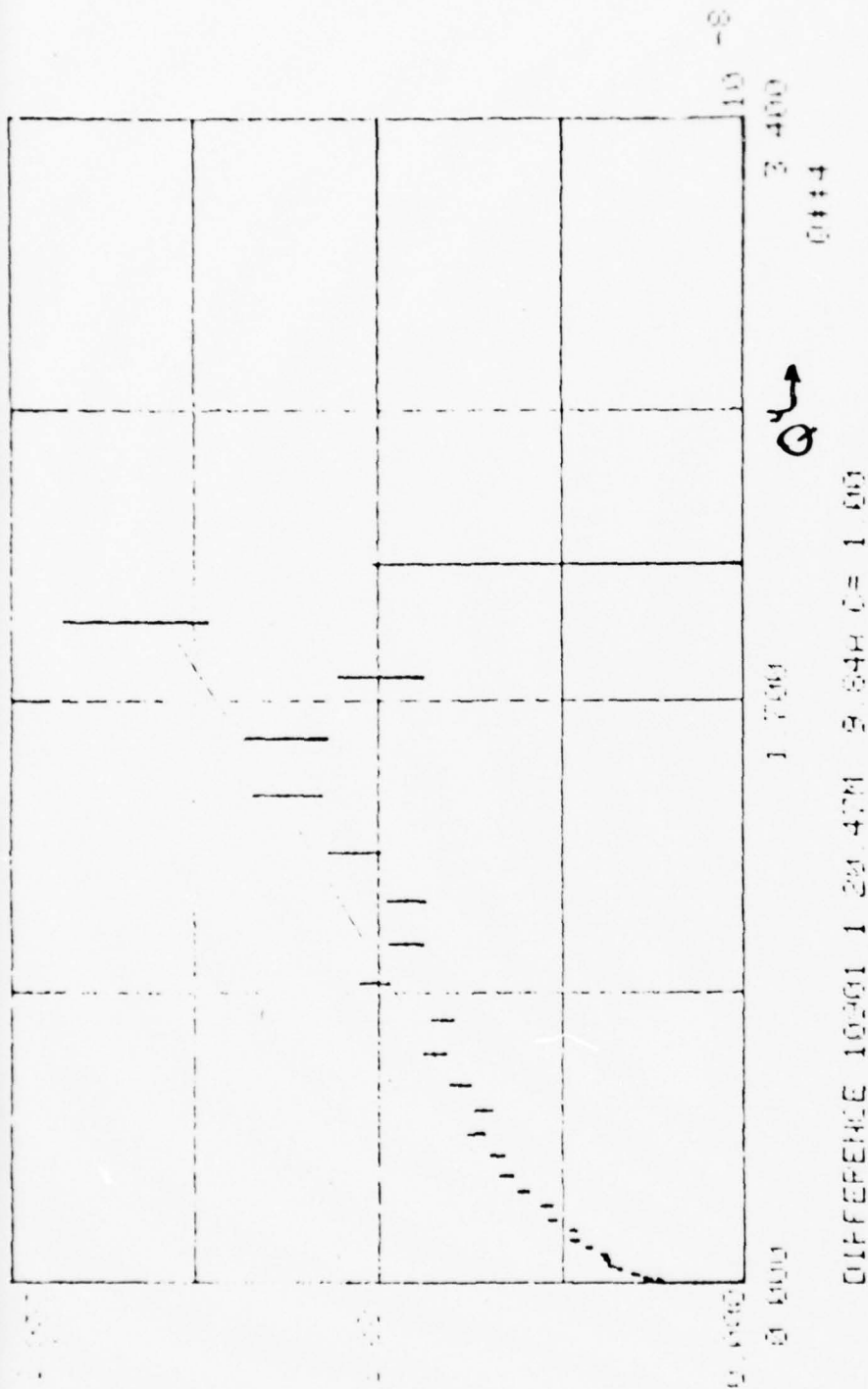


Figure 5b

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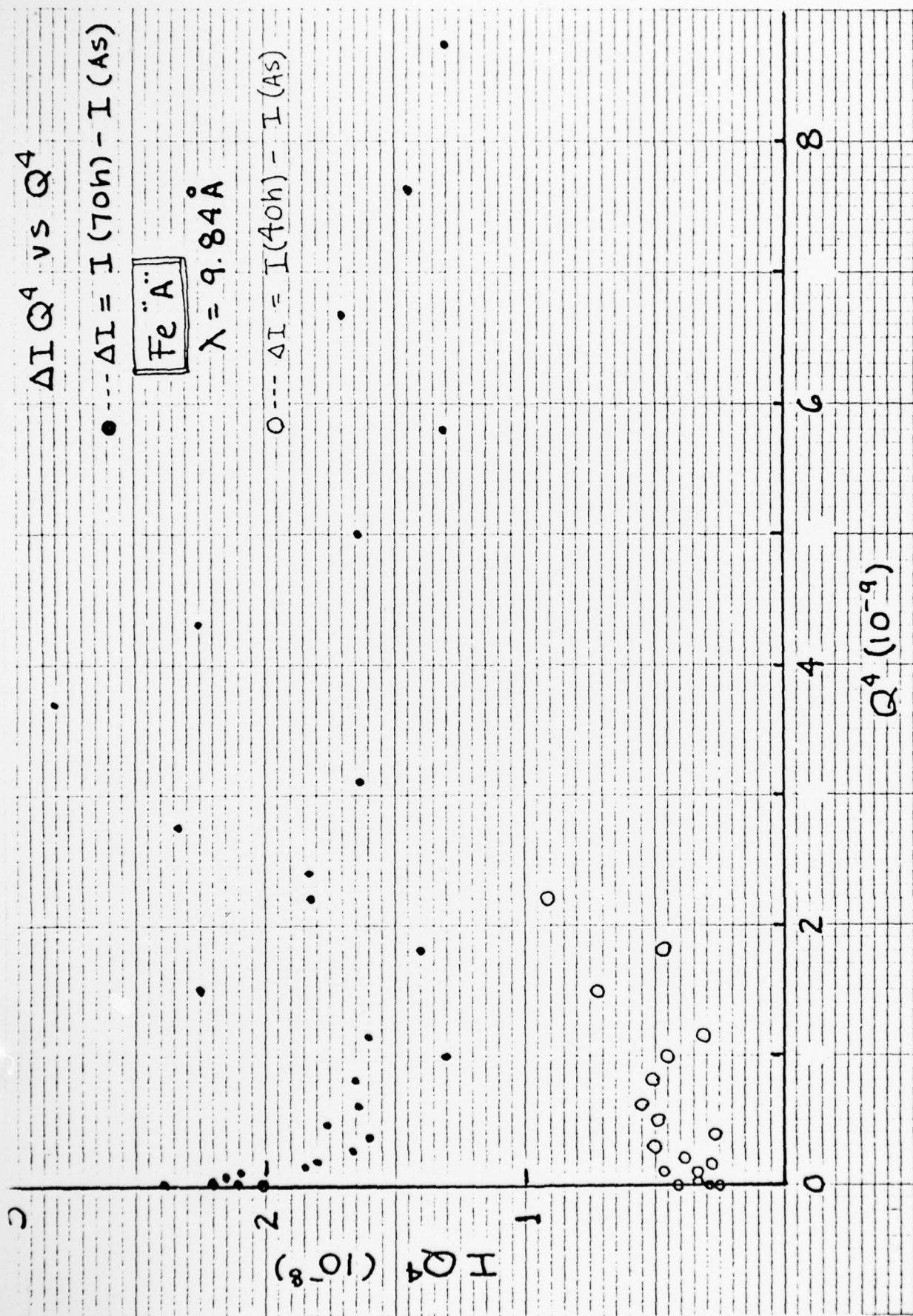


Figure 6a

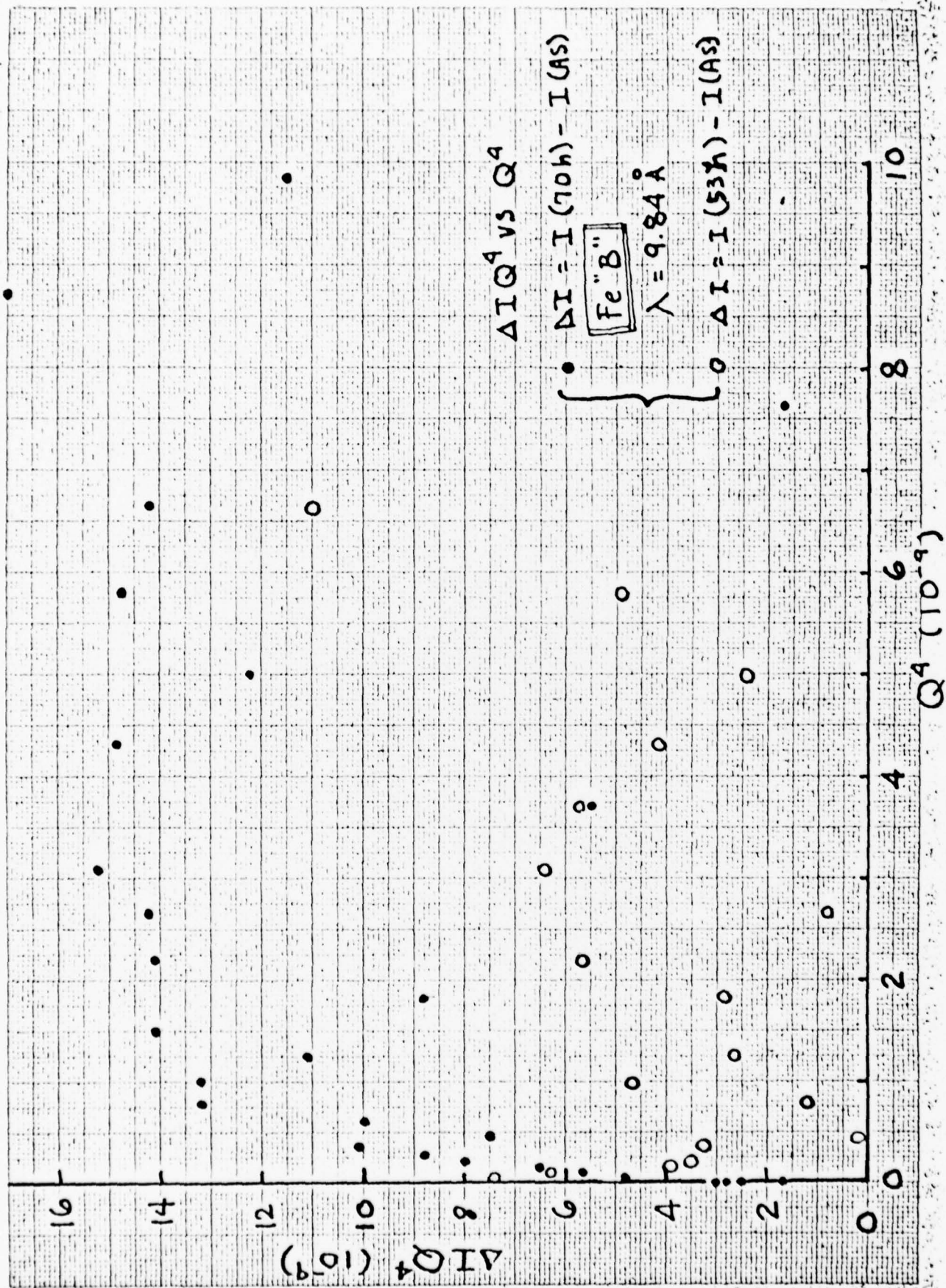


Figure 6b

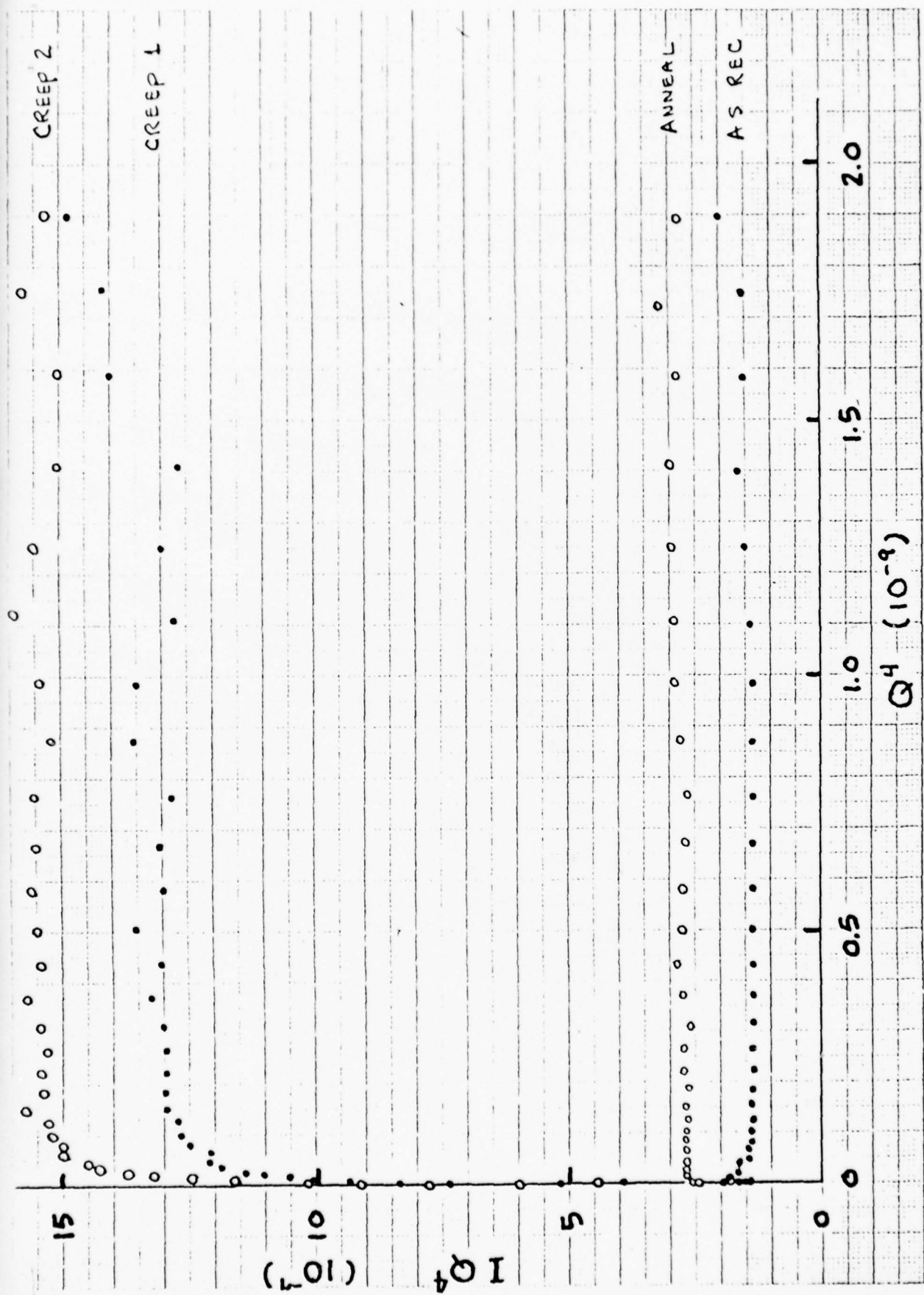


Figure 7a

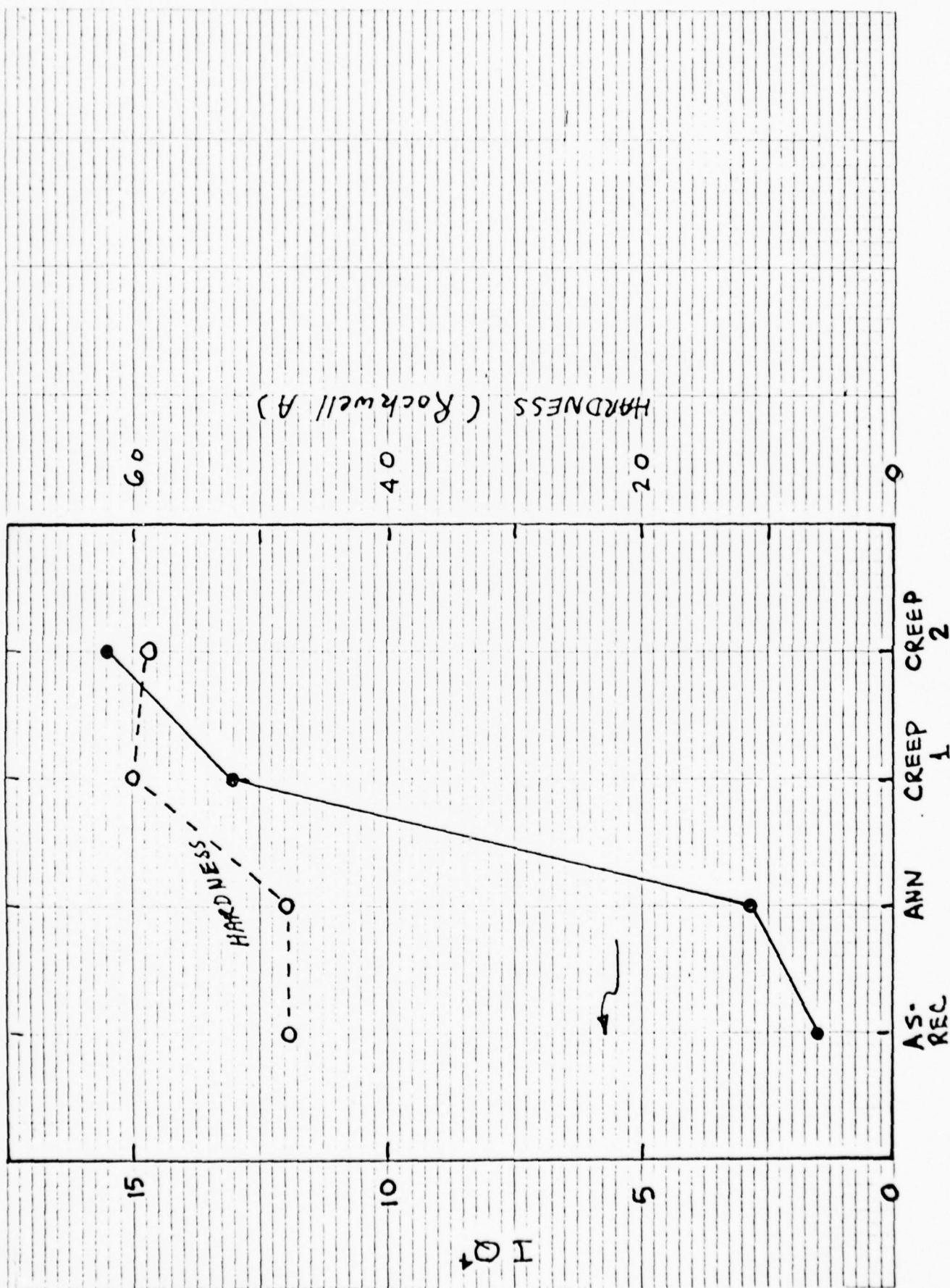
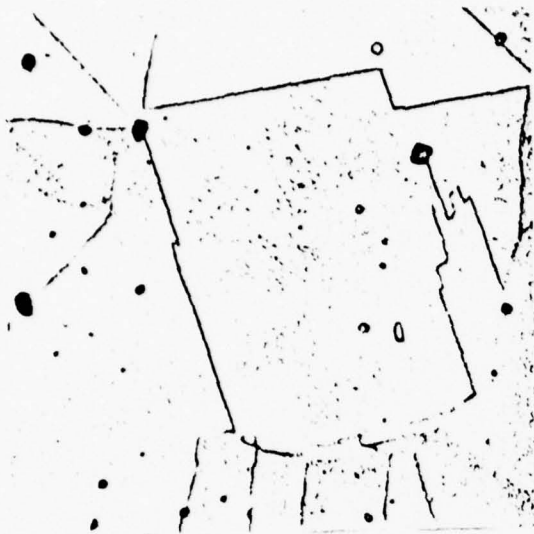
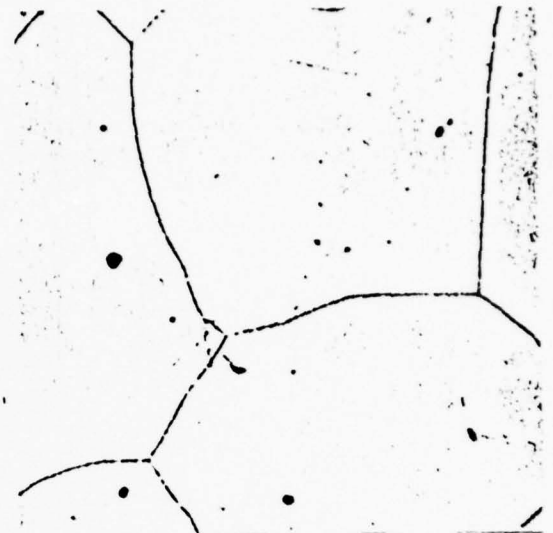


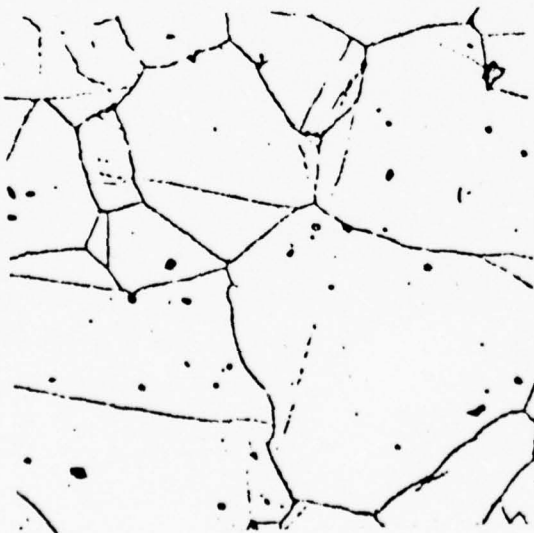
Figure 7b



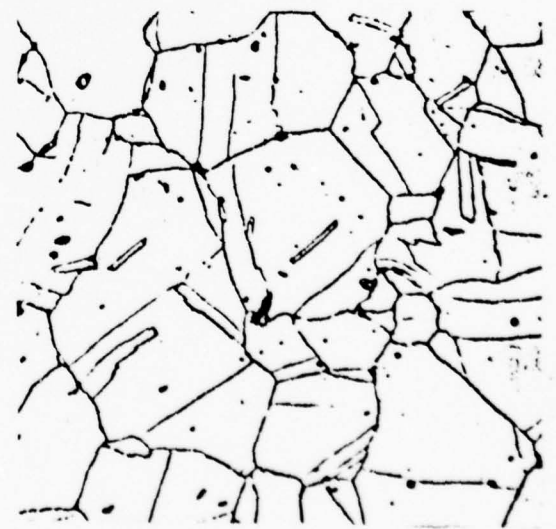
As-received



Annealed

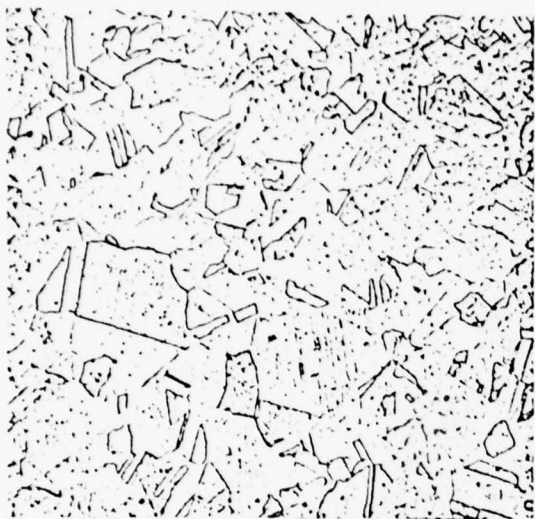


Creep 1

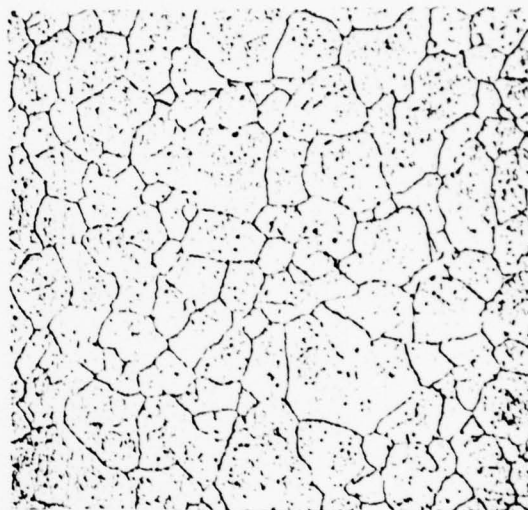


Creep 2

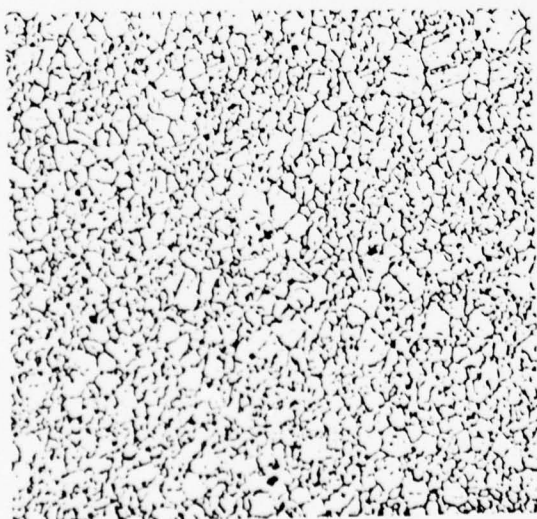
Figure 7c (1000X)



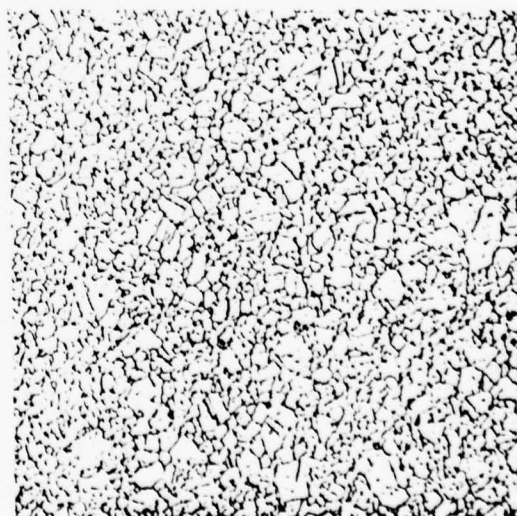
As-received



Annealed



Creep 1



Creep 2

Figure 7d (100X)

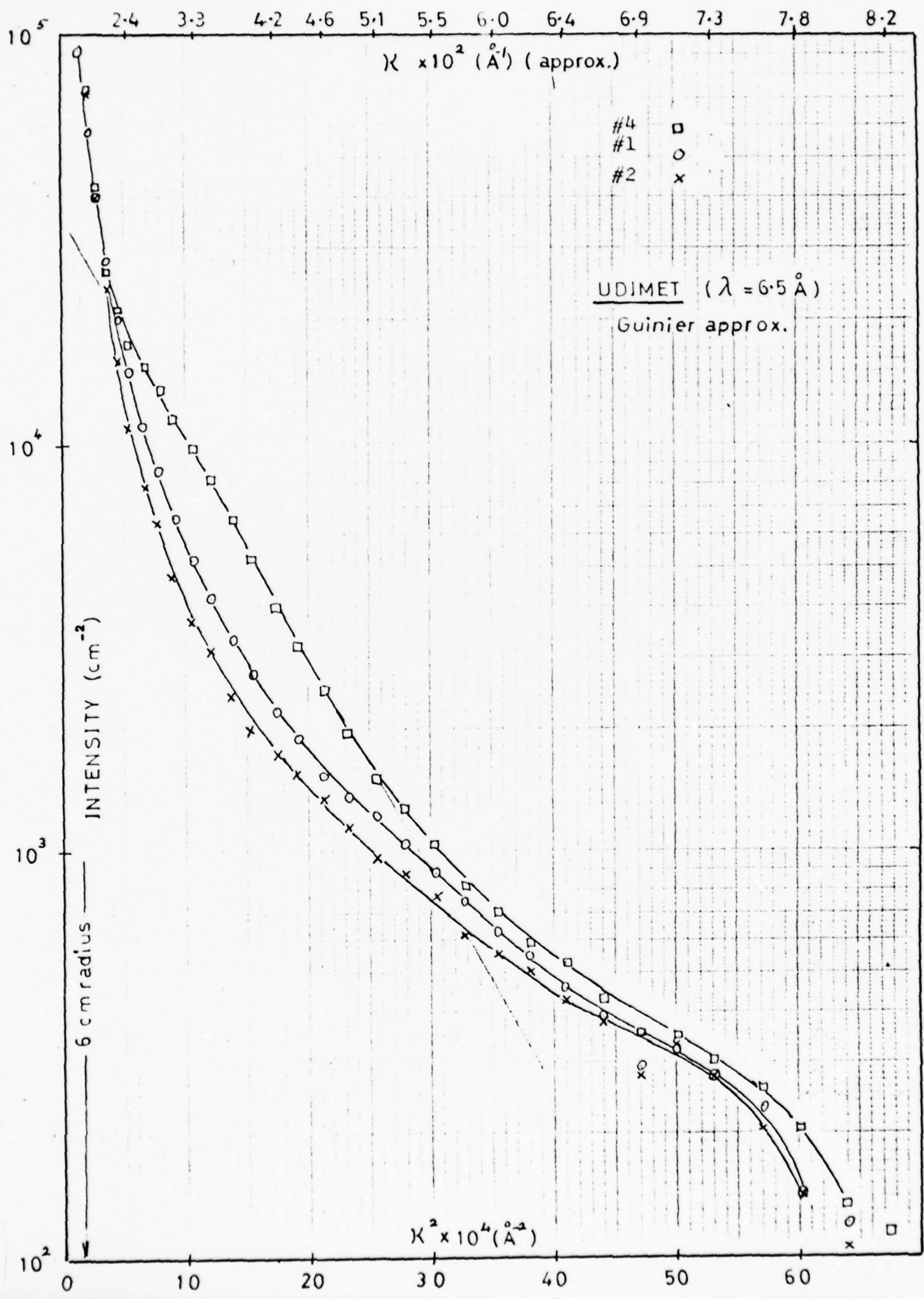


Figure 8a

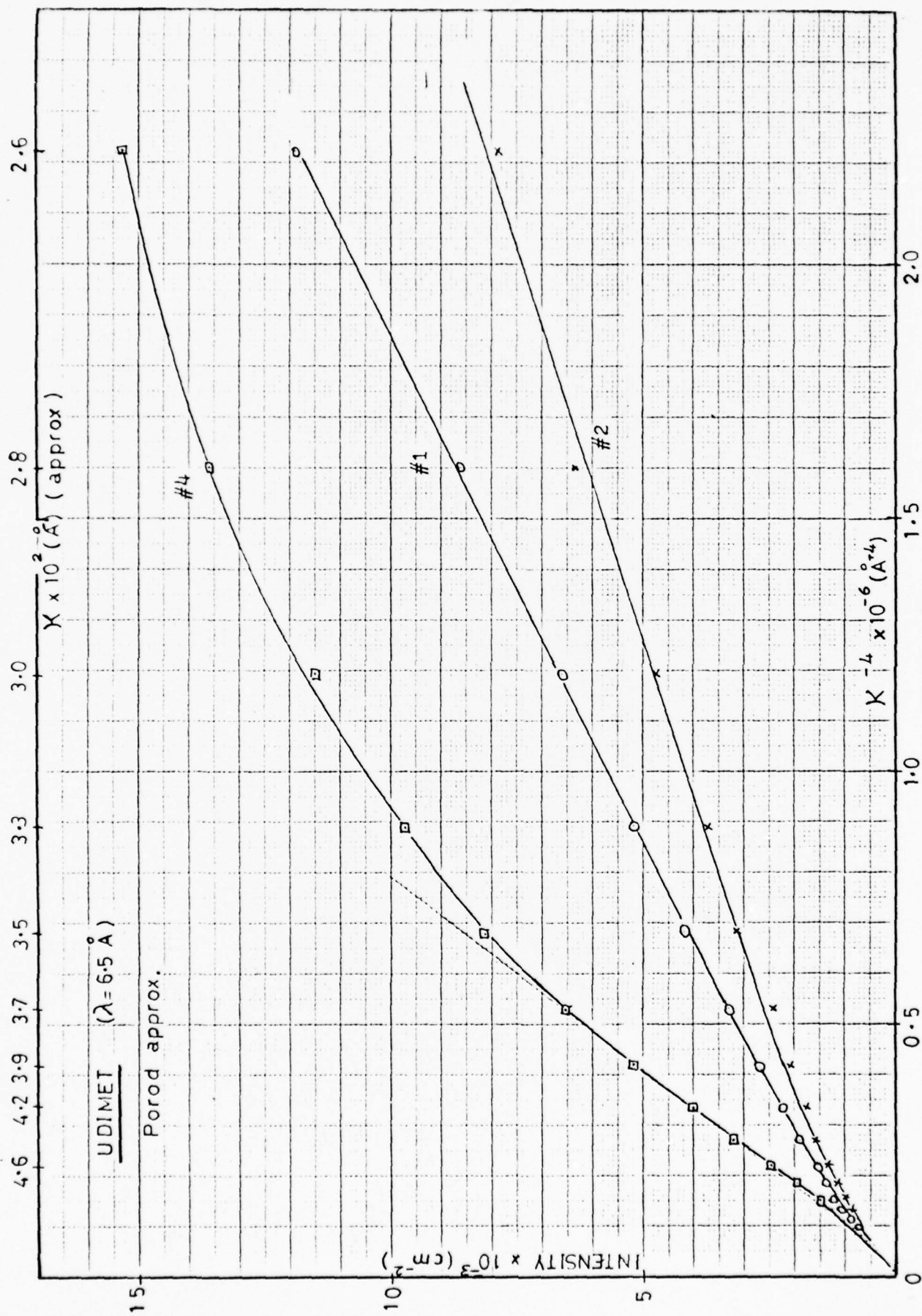


Figure 8b

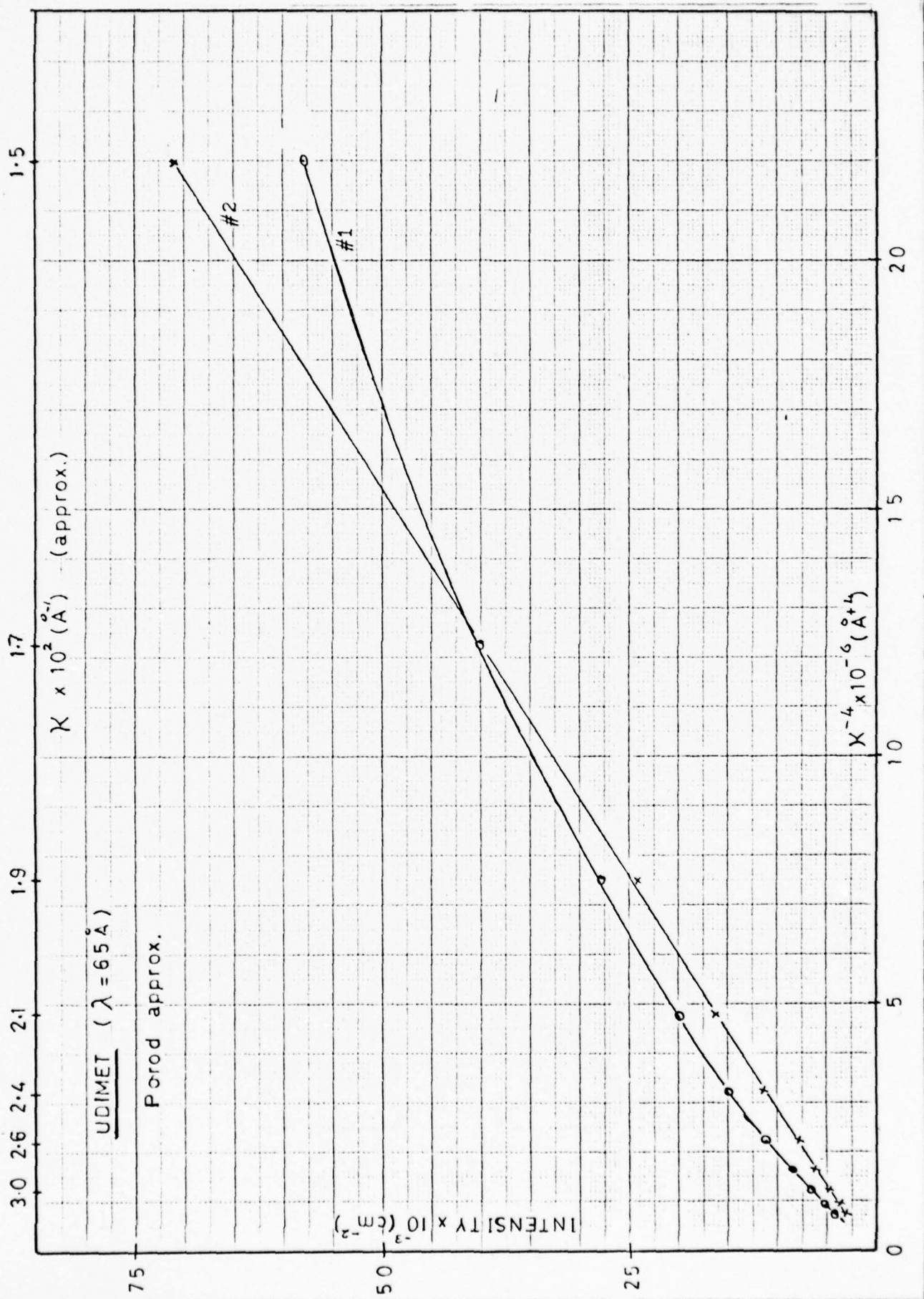


Figure 8c

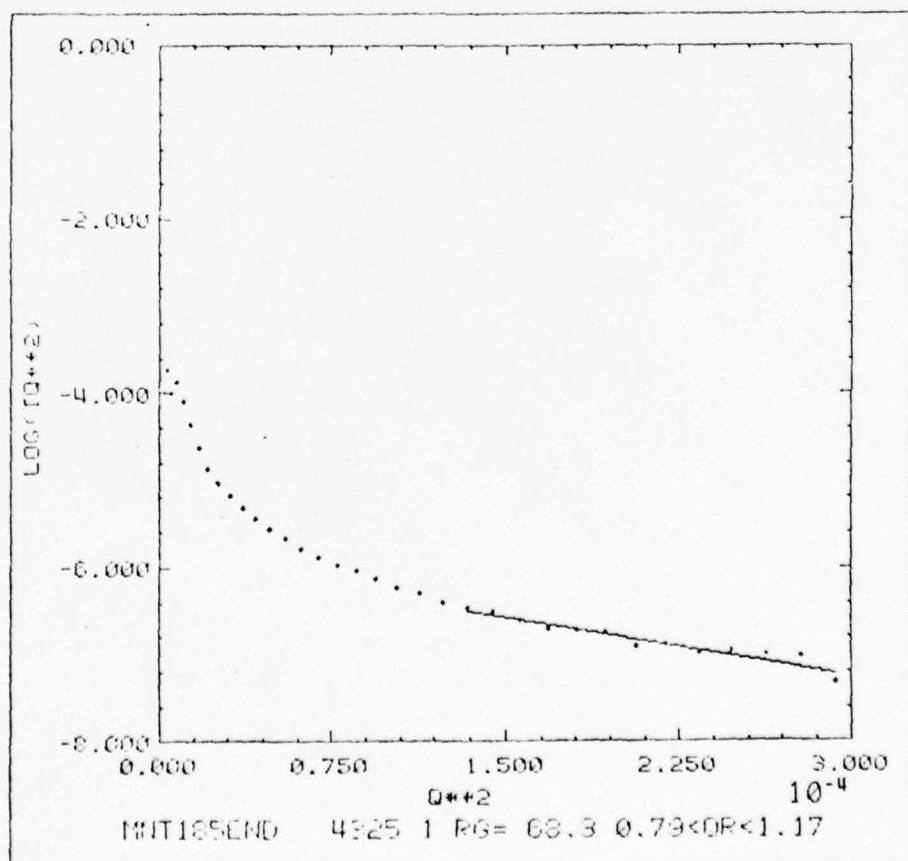
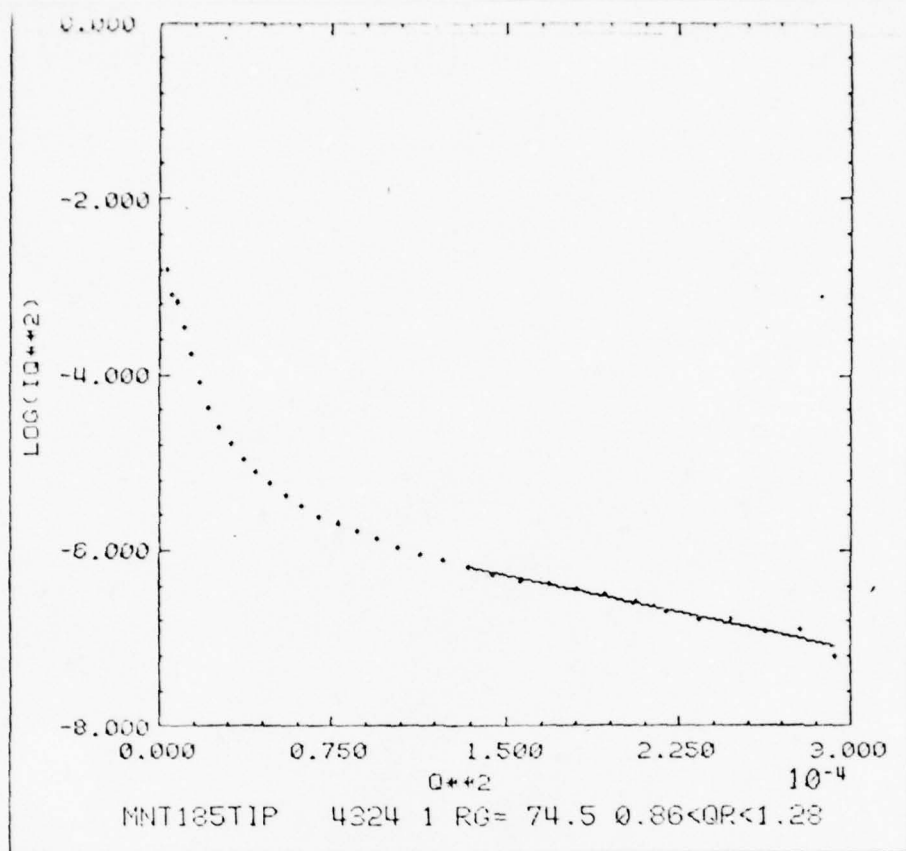
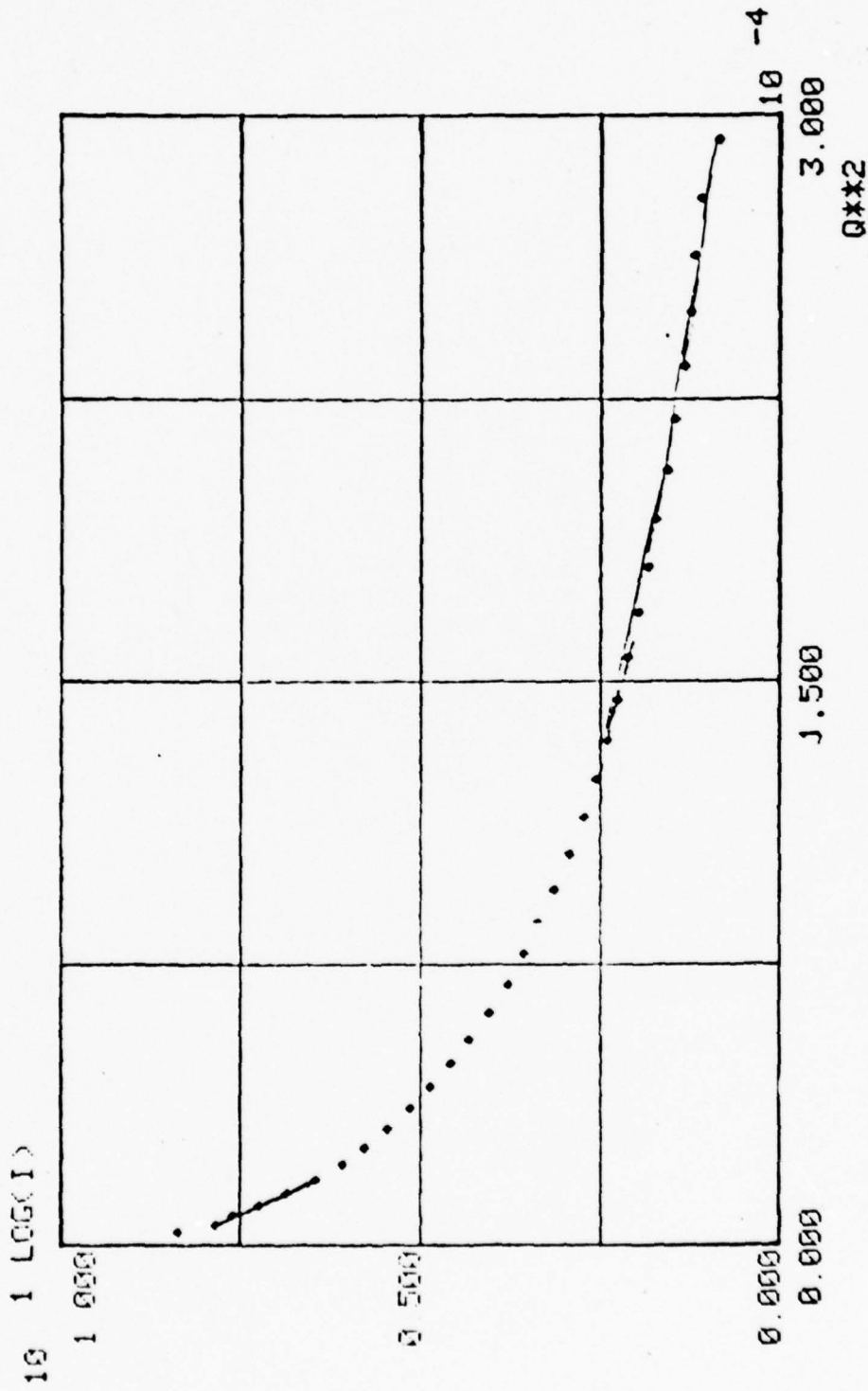


Figure 9a

$\gamma = \text{MX} + \text{C}$: $\text{M} = -0.1165\text{E}+06$ $\text{DM} = 0.29\text{E}+04$ $\text{C} = 8.473$ $\text{DC} = 0.35\text{E}-01$
 INTENSITY/CONC. ($Q=0$) = 4793. ERROR = 0.17E+03
 RADIUS OF GYRATION = 591.1 ERROR = 7.3
 $0.23\text{E}-02 < Q < 0.42\text{E}-02$ $1.37 < QR < 2.46$
 JEPSHTEC OUTPUT ? (Y) :



INC800H,ST 4342 1 RG=591.1 1.37<QR<2.46

Figure 9b

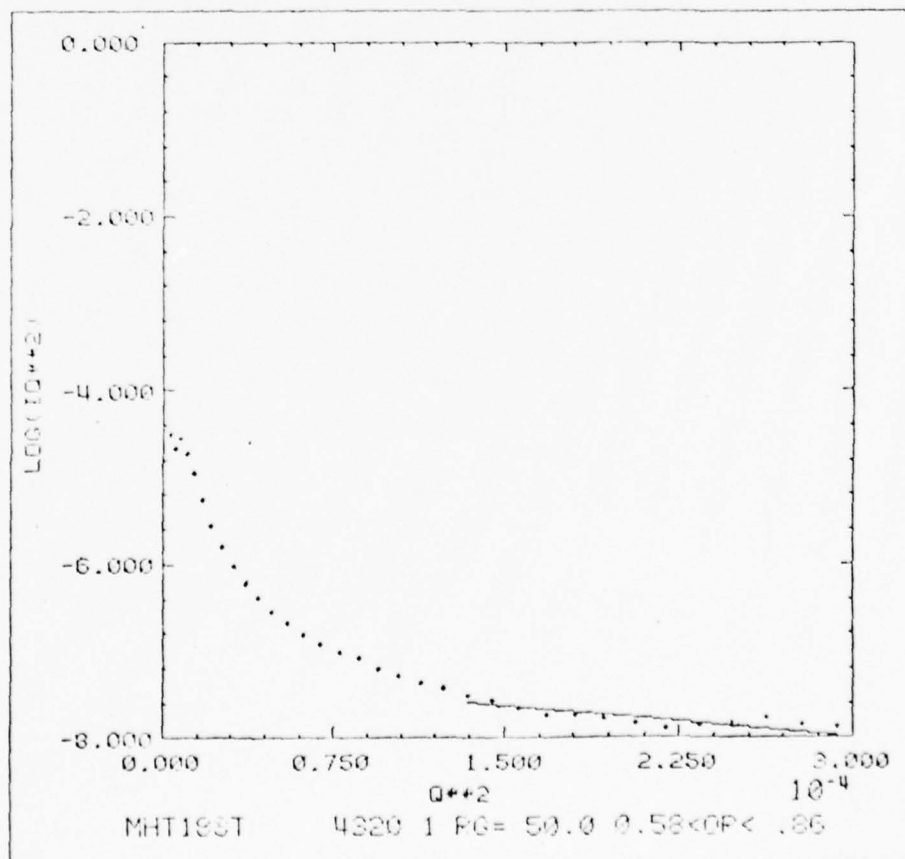
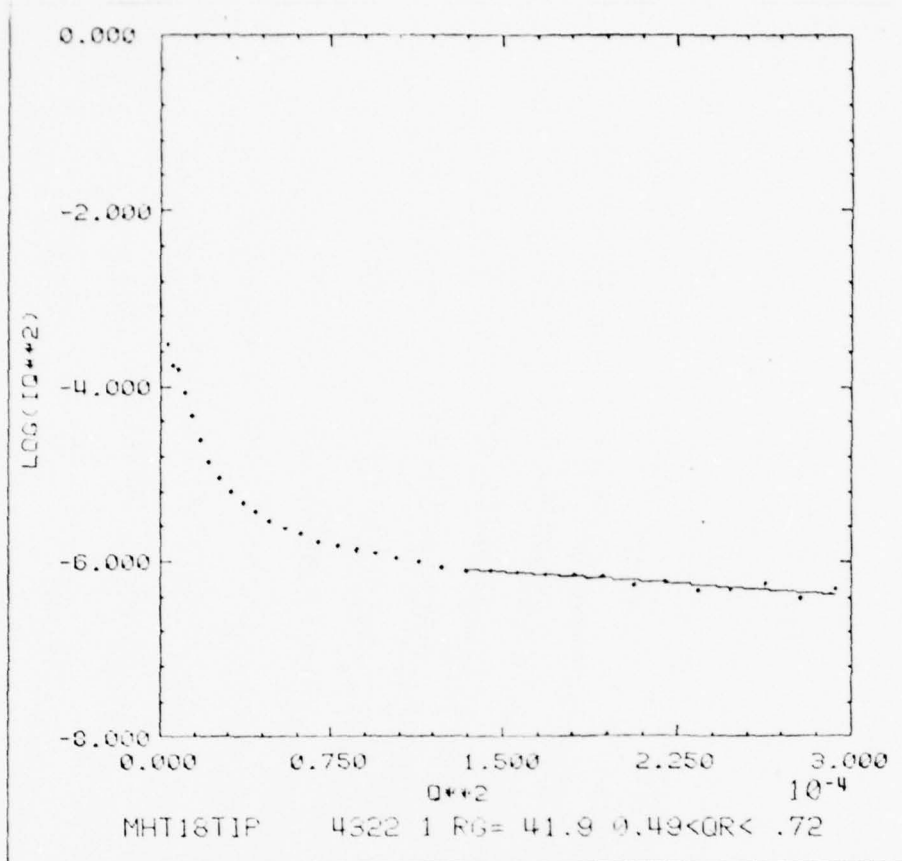


Figure 10

References

1. "Non-Destructive Evaluation of Materials with Cold Neutron Beams", NAVAIR Contract No. N00019-77-M-0418 (Dec. 1977); H. Herman.
2. Progress Report - "Feasibility Study on the Use of SANS for Microstructural Determinations of Technological Alloys", Contract No. N00014-78-M-0074 (Aug. 1978); H. Herman.

Appendix 1

Progress Report

to

Office of Naval Research
Arlington, Virginia 22217

by

Professor Herbert Herman
State University of New York
Stony Brook, N.Y. 11794
(Mailing Address - 11 Meroke Trail
Port Jefferson, N.Y. 11777)

Contract No. N00014-78-M-0074

Feasibility Study on the Use of
Small-Angle Neutron Scattering for
Microstructural Determinations of Technological Alloys,
Carried out at Atomic Weapons
Research Establishment (AWRE),
Aldermaston, England

Progress Report

SANS Study of Technological Alloys at AWRE

Alloys which were (and are currently being) studied:

1. Titanium Alloy from Disc Section-Fatigue (Ti-6-4).
2. HY-130; Hydrogen Embrittlement of Notched Specimen.
3. Stainless Steel (Type-304) - creep study.
4. Pure Iron-Hydrogen Embrittlement.
5. UDIMET 700 - HIP; Heat treated and Aging study.
6. Incoloy.
7. Hastelloy.
8. Inconel.
9. Aluminum Oxide - (Alumina) - Prestressed and Sintered Varying Densities.

This study, which is aimed at showing feasibility, was carried out by this investigator at "The Atomic Weapons Research Establishment", Aldermaston, England (AWRE). AWRE is a laboratory administered by the Ministry of Defense, U. K.

This preliminary research program on non-destructive evaluation using small-angle neutron scattering (SANS), was initiated and supported by the Naval Air Systems Command and the Office of Naval Research. The central goal of this aspect of the effort was to show feasibility of SANS through studies of technological alloys in conjunction and at AWRE. This investigator spent the month of July working together with Dr. R. Miller and his associates on the HERALD REACTOR. In a previous report written by this investigator, (Non-Destructive Evaluation of Materials with Cold Neutron Beams - Contract No. N00019-77-M-0418 - December, 1977), the details of SANS and the AWRE facility were described. Since that time, a LETI area detector was installed and tested. In the main, the computer hardware has been installed, and software programs are operational, with only minor debugging being required. During this investigator's stay at AWRE, there were minor problems which were encountered: eg., absence of an interface between the data acquisition system and the data analysis computer. This

necessitated the transfer of data by paper tape which was mainly inefficient and annoying, but not really an essential problem.

Overall, during my stay, the computer systems operated ably and the reactor and cold source worked perfectly. There were no shutdowns following the original reactor startup cycle.

Since HERALD is limited to 5 megawatts, the flux output was low, but an experiment could be carried out in 2 hours for a strongly scattering system. Thus, a large number of experiments could be carried out during this investigator's visit to AWRE.

Dr. Miller will continue a number of the runs which were not completed. These results will be reported in the Final Report on this contract.

Below are given the experimental runs which were carried out and those currently underway. The details of these experiments and the findings will be given in the Final Report.

The range of experiments which were carried out and which are still undergoing study-to-completion will be presented according to the following scheme:

- I. Experiment
 - A. Alloy
 - B. Contact
 - C. Type of experiments performed
 - D. Results

I. Fatigue of Ti-Alloy Disk (Webs and rims)

- A. Ti-6V-4Al.
- B. Dr. G. London, NAVAIR.

C. SANS at central reduced section and at non-loaded end. A large number of experiments were performed at AWRE. In addition, Dr. M. Suenaga of Brookhaven National Laboratory and Dr. G. Kostorz of Max-Planck-Institut, Stuttgart carried out measurements on one of these specimens at ILL, Grenoble, France, at longer wavelengths and at considerably greater neutron fluxes.

D. An increase has been detected in scattered intensity which is attributable to fatigue. Theoretical and experimental studies were carried out to evaluate double-Bragg scattering due to the $Ti_3 Al (\alpha')$ phase.

In addition to SANS studies, cold neutron radiography (CNR) was studied of these specimens. In one very interesting case, the utility of CNR was demonstrated when the full extent of a crack was observed with CNR; with normal thermal neutron radiography, the crack was barely noticeable. The plates will be included in the final report of this study.

II. Hydrogen Embrittlement of HY-130

A. HY-130 steel

B. Mr. Zannis, NSRDC, Annapolis, Maryland.

C. To be completed - in situ
charging of notched fracture mechanics specimens.

III. Hydrogen Embrittlement of Iron

A. Pure Iron

B. Dr. M. Suenaga, Brookhaven National Laboratory

C. Hydrogen introduced at 475°C under pressure of 900 psi
for up to 70 hours. Methane bubbles presumed to have formed

D. Clear scattering, dependent on amount of hydrogen, at Porod
region due to voids

IV. Creep of Stainless Steel

- A. Type 304 stainless steel
- B. Dr. M. Suenaga, Brookhaven National Laboratory
- C. Creep of 304 stainless steel at 575°C - interruption experiment
- D. Significantly increased scattering at the Porod region (higher angle portion of the SANS regime), clearly indicative of void formation. Effect is dependent on time of creep.

V. Hot-Isostatic Pressed Superalloy

- A. UDIMET 700
- B. Dr. V. Wilms, MTU, Munich
- C. Heat treated and ageing of HIPPID UDIMET 700
- D. SANS shows excellent results for the aged specimens, the small second phase, 70 Å, being clearly identifiable and comparable with accepted TEM results.

VI. Miscellaneous Superalloys

Incoloy, Hastelloy, HIPPED alloys, Inconel (creep failure specimen) are currently under study at AWRE. Some preliminary results indicate that the scattering is strong. The results will be included in the final report.

VII. Pressed and Sintered Pure Al_2O_3

These specimens are available to us in densities ranging from ~60% - ~100% of theoretical density. The specimens scatter very strongly due to large internal surface areas. The final results will be contained in the final report.

Conclusions

At this stage it can be concluded that SANS represents a good pos-

sibility for an effective method of metallographic analysis. The technique affords the possibility of evaluating average bulk effects in both two phase precipitation strengthenable alloys and in void-containing systems. It is clear to us that long wavelength neutrons are essential for most of the technological work which we encountered, i.e. a cold source.

The above experiments were carried out principally at AWRE, Aldermaston, but several were also carried out at ILL, Grenoble.

The final report (due by October 31, 1978) will detail the results.

The primary purpose of a small-angle scattering experiment is to determine the angular distribution of the scattered intensity. Methods of analysis of these data provide information about parameters such as size, shape and distribution of the inhomogeneities.

Radius of Gyration

The intensity of small-angle scattering is related to the differences in coherent scattering length density between the particle and the matrix as well as to the number of scattering centers. In the case of groups of widely separated particles (dilute concentrations) which are randomly oriented, the scattered intensity is given by

$$I(Q) = I_n(Q) N F^2(Q) \quad (1)$$

where N is the average number of scattering particles, $I_n(Q)$ is the intensity scattered by a single nucleus and $F^2(Q)$ is the average of the square of the structure factor of the particle and is also the Fourier transform of the shape of the particle. The quantity Q is given by

$$Q = \frac{4\pi \sin \theta}{\lambda} \quad (2)$$

where λ is the wavelength of the incident neutrons and 2θ is the scattering angle.

*Reference 1 should be sought for a more thorough review of this field.

The scattering intensity at very small angles can be closely approximated by

$$I(Q) = I_n(Q) N n^2 \exp[-Q^2 R_g^2/3] \quad (3)$$

where n is the number of nuclei per particle and R_g is the radius of gyration of the particles. Although strictly valid at zero scattering angle (for $Q R_g \lesssim 1.5$), Eq. (3) is a sufficiently good approximation to represent the intensity curves over a finite region, especially for spherical particles. Therefore, the radius of gyration can be determined from the slope in the linear region near zero angle on a plot of $\log I(Q)$ -vs- Q^2 . From Eq. (3), when θ is measured in radians, we obtain for the radius of gyration:

$$R_g = \frac{\lambda}{4\pi} \left| \frac{3(-\text{Slope})}{\log \theta} \right|^{1/2} \quad (4)$$

R_g is related to actual radius, R , for a set of spherical particles by

$$R = \sqrt{\frac{5}{3}} R_g \quad (5)$$

At sufficiently large angles (i.e., $R_g Q \gtrsim 2.5$), the Guinier approximation is no longer strictly valid.

The above procedure holds for a system of widely separated and randomly oriented particles. If the concentration of scattering particles is high or if their interaction is strong, the curve of $F^2(Q)$, will be modified, and the apparent R_g , as obtained from the Guinier plot, will be smaller

depends only on the total volume of the dispersed phase, and is independent of the degree of dispersion; that is, the volume of the particle.

The zero value of intensity, $I(0)$, increases, for a fixed concentration, with the volume of the particles, as seen from,

$$I(0) = I_n(0)NV^2\rho^2 \quad (7)$$

V is the volume of the particle and, ρ , its nuclear density. For a fixed concentration, NV and ρ are constant, and $I(0)$ increases with V . Hence, the quotient $I(0)/Q_i$ is a measure of the volume V of the particle. It can be shown that

$$\frac{I(0)}{Q_i} = \frac{V}{2\pi^2} \quad (8)$$

Thus, no absolute intensity measurements need be made to evaluate the volume of the particle.

surface area: Porod has shown that for the special case of a two-phase system, each phase being of constant nuclear density and arbitrarily distributed,

$$\lim_{Q \rightarrow \infty} I(Q) = 2\pi I_n(Q)(\Delta\rho)^2 S Q^{-4} \quad (9)$$

where $\Delta\rho$ is the difference in nuclear density between phases 1 and 2 and S is the total area of the interface separating the phases. The absolute value of intensity at large Q , i.e., the higher angle region of the diffraction curve, depends then on only $\Delta\rho$ and S . The equation predicts

that the outer portion of the scattering curve decreases asymptotically with Q^{-4} . This fact is useful in confirming the existence of a sharp interface between the heterogeneities and the matrix and for establishing the range of Q over which the surface area measurements can be validly carried out. This method involves the absolute measurement of $\lim_{Q \rightarrow \infty} I(Q)$ and since $\lim_{Q \rightarrow \infty} I(Q) \ll I_0$, the intensity for the incident beam, this cannot be done satisfactorily.

To avoid the difficult measurement of I_0 , Porod introduced a normalization procedure. He showed that S/V , the surface-to-volume ratio, known as the specific surface, S_{sp} , can be expressed as

$$S_{sp} = \pi C_1(1-C_1) \left[\lim_{Q \rightarrow \infty} I(Q) Q^4 \right] \left| \int_0^{\infty} Q^2 I(Q) dQ \right|^{-1} \quad (10)$$

where C_1 is the fraction of the volume occupied by the phase 1. If the particle volume, V , is known, the surface area can be determined. The difficulty is that the normalization integral must be evaluated between zero and infinity in Q , while the experimental data covers only a small part of this range. Extending the integrand to the upper limit can be done analytically, since the high angle part of the experimental data depends on Q^{-4} . However, extrapolating the data to zero angle is a more uncertain procedure, and, hence, there will always be some uncertainty as to the accuracy of the results.

The above methods of analysis have been applied to the study of shape, size, and distribution of precipitates in metallic systems, in particular, GP zones in age-hardened aluminum alloys. The distribution of the zones is polydisperse, and interparticle interference effects predominate. Hence,

the computed results must always be treated with caution. This will be
expecially true for much of the applied work which is discussed here.

Appendix 3

Contribution to ONR-London "European Scientific Notes";
Oct. 1978.

European Small-Angle Neutron Scattering - An Update

Small-angle scattering of light, x-rays and neutrons are techniques which can be used to study colloidal-like particles ranging in size from ten's to thousand's of Angstroms. A considerable amount can be found out about a number of average properties of such fine particle systems: e.g. size, size distribution, shape, surface characteristics, internal structure of biological and polymeric substances. The range of research fields which is utilizing small-angle scattering techniques is vast indeed: biology, physics, chemistry, materials science.

In more recent years, small-angle neutron scattering (SANS) has emerged as a particularly important technique in the realm of "fine particle" science. In fact, SANS has developed principally in Europe. X-rays and optical small-angle scattering have also developed strongly, but SANS is the fair-haired technique of the day. In the not-so-distant future, with the emergence of storage rings (e.g., the National Synchrotron Light Source at Brookhaven National Laboratory), where intensity and collimation will be increased significantly, x-ray small-angle scattering will undoubtedly once again be rediscovered (ESN-30-8, p. 367-369). But for the time being, neutrons are opening doors which were once previously closed for a large number of experiments.

This writer has in the past presented a number of views of the SANS situation in Europe (ESN-30-1, p. 36-37; ESN-30-2, p. 73-76; ONRL 17-75, "Small Angle Scattering at Jülich, West Germany). In the two or so years since those reports were published, SANS has continued to grow in the UK and on the continent, where a range of new instruments have been built or are being planned. I would here like to bring things up-to-date with

respect to SANS in Europe.

Before doing so, it would be best to review those properties of neutrons which make SANS so desirable a technique. The neutron, for starters, can penetrate a thicker specimen, so that, for example, large metallurgical specimens can be examined. Of further importance, neutrons having wavelengths greater than say 10 \AA can be employed, still with good transmission, and can thus be used to avoid double-Bragg scattering from crystalline specimens (see below). Another important aspect of SANS is the possibility of the occurrence of a considerable differential scattering length between two nuclei, when, for the case of x-rays, no intensity might be forthcoming (e.g., Al-Mg alloys).

The SANS prototype, and the leader as well, is the Institut Laue-Langevin at Grenoble, France. ILL remains the de facto international center of the field, with workers coming from the world over to carry out SANS experiments at this unique facility. ILL is administered by a tripartite international group comprised of Britain, France and Germany (the directorship rotates among these three). Candidate experiments are submitted by scientists from these three countries and are reviewed by the Scientific Committee, who judge the proposal's relative merits. If accepted by the Committee, a specified number of days (e.g., one through four) are allotted. Experimenters receive assistance from resident scientists, who are frequently directly involved with the programs. Nationals outside of the tripartite do use ILL, but access under such circumstances will be through an interested, and sympathetic, resident scientist.

There are complaints that the ILL system is too rushed and that it is most difficult to carry out good, thoughtful science in a stop-clock environment. Indeed, anyone who has experienced the fatigue of round-the-clock days

at ILL will appreciate this view. On the other hand, the facility is highly automated, the computer hardware being such that good experiments can be done quickly -- if (!) proper care is taken in experimental design.

The central criteria for state-of-the-art SANS include: (1) a cold source (e.g., liquid H_2/D_2 within the reactor), so that the neutron energy spectrum can be shifted to lower values, i.e., longer wavelengths; (2) area detection, so that high sensitivity is achieved, in addition to enabling the detection of anisotropic scattering. For studies of crystals, if neutron wavelengths can be used which are about twice the Bragg cut-off, doubly-scattered radiation can be avoided, thus, eliminating annoying parasitic background in studies of poorly-scattering systems (e.g., defects in crystals). In addition, the reactor must be of adequate power to yield intensities sufficiently great to give meaningful scattering above background. Even a 5MW reactor is capable of this.

ILL is eminently able to yield optimum scattering conditions. This, together with fine hardware and a highly professional staff, make the Grenoble facility outstanding. It should be pointed out that a tremendous number of different kinds of experiments are carried out at ILL in biology, polymers, physical chemistry, physics, metallurgy, etc.

There are other extremely active European machines. The reactor facility at KFA, Julich, West Germany, developed and run by W. Schmatz, presently at Karlsruhe, has been highly productive in solid state physics studies. Much work has been done at KFA on defects, magnetism, superconductivity, and phase separation in alloys. The Germans who are associated with the KFA reactor, however, not infrequently take time at ILL due to the latter's superior intensity and the ability one has to go to lower scattering angles (the DII-SANS spectrometer has a total length of 80 meters!).

In France, at Saclay, the DIDO reactor now has a cold source and a LETI detector. This facility is engaged mainly in biological and polymer studies. And at Orphay, in France, a new medium flux reactor is under construction, with an expected completion date of 1980. Several SANS spectrometers are being considered for the Orphay reactor, and a cold source is planned as well.

In Italy, there is a SANS facility on the 5MW CAMEN reactor in Pisa. This reactor, which belongs to the Italian Army, is rented in part by FIAT, which is carrying out non-destructive testing using SANS. Specifically, P. Pizzi and H. Walther employ a novel SANS spectrometer, balanced by wires and counter-weights to adjust for the effects of sinking, Leaning Tower style. Their cold source is liquid propane, and they use a LETI area detector. Pizzi and Walther have been remarkably productive with this less-than-ideal set-up. They have, in fact, pioneered the field of NDT using SANS. It is also rumored that Ispra will provide hardware for still another European SANS laboratory. Things are still in the talking stage, but considered is a spallation unit for the generation of neutrons. Scientists in Germany, by the way, are in pursuit of funds for the construction of a pulsed SANS system. No details yet as to location.

Meanwhile, the UK, though having the availability of ILL, are, too, building with some apparent vigor.

Fully operational, and now spewing out data, is the 5MW HERALD Reactor at the Atomic Weapons Research Establishment (AWRE), Aldermaston, England, which is close to Reading and not far from Harwell. The reactor is in a "sanitized" part of this secret defense laboratory. The AWRE-SANS spectrometer, fitted out with a H_2/D_2 cold source and a recently installed LETI

detector, is administered by Robin Miller, who, together with Roger Stewart of the Physics Department, University of Reading, are looking at "applied" SANS. Specifically, they are examining creep of Nimonic alloys. A number of groups use the AWRE machine to study polymers and metal alloys. The HERALD SANS facility is a fine, small unit, which is easy to run, and a delight to behold.

Another facility at HERALD, but not directly related to SANS, is a Cold Neutron Radiography (CNR) development program, under the guidance of G.S.G. Tuckey, who is doing truly remarkable things with CNR. The resolution and contrast are exceedingly high, and present new schemes of neutron radiography (as well as a further justification for a cold source!).

Further in the UK is the Harwell PLUTO Reactor, which now has a 3 meter SANS spectrometer and a just recently installed LETI area detector. There is no cold source on PLUTO, so their capabilities will be limited.

Also at Harwell, but perhaps more interesting, is the current construction of a LINAC, which should be on line in early 1979. The machine is being built for the UK's fusion program, but clearly high fluxes of neutrons will be forthcoming. It is planned to develop a position sensitive scintillation detector, with fiber optics leading to photomultipliers. This unit will be designed and constructed at the Rutherford Laboratory. It is argued that this machine will prove the usefulness of pulsed neutron beams.

There are currently four SANS units which can be used by UK scientists: at ILL there is D11 and D17 (a newly constructed SANS spectrometer); PLUTO (Harwell); and HERALD (AWRE). Roger Stewart (Reading) claims that a SANS machine is needed offering a large and variable vector range up to 1 \AA^{-1} . He also argues for the possibility of separating the elastic and inelastic scattering, with good spatial resolution. Stewart states: "An additional

factor which must not be overlooked is that the proposed machine....will be in the UK with a flux comparable to D11. This has many advantages for UK scientists; ease of access, no customs problems for samples and equipment, more immediate studies of perishable samples, etc."

Stewart and his co-workers have spent considerable time considering pulsed neutron sources. Care must be taken, he argues, not to design-out flexibility from such a unit. For example, he is concerned with designing too small a beam cross-section.

All in all, Stewart's arguments for pulsed sources are attractive and will demand our future attention.

It is clear that the Europeans have led the world in SANS. Certainly there is much more to be done. In recognition of this, in the United States, the National Science Foundation has awarded funds to Oak Ridge National Laboratory to develop a user-oriented SANS resource. This is indeed gratifying, but comes some 5 years after the ILL-SANS facility was an operational fact. We are, thus, somewhat behind the Europeans. But an added problem is that the Oak Ridge machine will have no cold source, limiting applications, in the main, to non-crystalline systems. This represents a serious shortcoming. American scientists, in need of state-of-the-art SANS, will have to keep traveling to Europe for the foreseeable future.

However, there are some recent activities underway, in that Argonne National Laboratory and Los Alamos are considering various accelerator-generated neutrons for SANS. And the National Bureau of Standards has a SANS spectrometer, also without a cold source.

SANS is truly an interdisciplinary experimental technique. Its range of applications to the life sciences, the physical sciences and to NDT

-7-

studies is awesome.

The Europeans have shown us once again, as with automobiles, that small can be beautiful.

H. Herman
Dept. of Materials Science & Engineering
State University of New York
Stony Brook, N.Y. 11794